To my parents

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An De Schrijver

ACIDIFICATION AND EUTROPHICATION OF FORESTS ON SANDY SOIL: EFFECTS OF FOREST TYPE AND DEPOSITION LOAD

Thesis submitted in fulfillment of the requirements

for the degree of Doctor (PhD) in Applied Biological Sciences

Verzuring en vermesting van bossen op zandbodem: effecten van bostype en depositielast

Illustrations on the cover:

Mixed stands of pine and oak at Mattemburgh (Pictures: Robbie Goris)

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A team effort

When Lance Armstrong won his fourth Tour de France, he not only followed tradition by dividing the prize money among his team-mates, he also doubled the amount to his fellow cyclists out of his own pocket. This largesse recognized that without his team's support, Armstrong wouldn't have won the race. It also demonstrates best practice in team science – rewarding the supporting cast that makes a lead author's findings possible, rather than hoarding all of the credit for oneself.

Nature (2003), volume 424, p. 471.

Het team:

Luc De Keersmaeker die de eerste steen legde van het biogeochemisch bosomvormingsonderzoek.

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Ten slotte nog dit:

De krant De Standaard kopte op 13 mei 2003 op de voorpagina: 'Zure regen bestaat niet' en 'Bossen zijn belangrijke bronnen van vervuiling'. Ik hoop dat dit werk helpt het tegendeel te bewijzen.

Michelbeke, April 2007.

Contents

List of abbreviations and chemical compounds

Abbreviations

BD	Bulk precipitation deposition
CE	Canopy exchange
CL	Canopy leaching (CE > 0)
CRM	Standard reference material
CU	Canopy uptake (CE < 0)
CV	Coefficient of variation (%)
DD	Dry deposition
DDF	Dry deposition factor
ICP	International Co-operative Programme on assessment and monitoring of air pollution effects on forests
LAI	Leaf area index (m^2 m^{-2})
NTW	Net throughfall water
P	Precipitation amount (mm)
S	Seepage flux of water (mm)
SD	Standard deviation
SF	Stemflow ion flux
TD	Total deposition = bulk precipitation deposition + dry deposition
TF	Throughfall water ion flux
TF + SF	Stand deposition of ions
BC	So-called 'base cations' or neutral cations (= $Na^+ + K^+ + Ca^{2+} + Mg^{2+}$)

Chemical compounds

Al^{3+} and Al(III)	Aluminium in soil solution can occur as Al^{3+} but also as several Al hydroxides. Therefore, when analyses of the soil solution are discussed, aluminium is presented as Al(III), when e.g. toxicity for roots is discussed, as Al^{3+}
Ca^{2+}	Calcium
Cl^-	Chloride
DON	Dissolved Organic Nitrogen
H^+	Proton
HNO_3	Nitric acid

K^+	Potassium
Na^+	Sodium
N	Nitrogen
NH_3	Ammonia
NH_4^+	Ammonium
NH_x	Reduced nitrogen (NH_3 and NH_4^+)
NO	Nitric oxide
N_2O	Nitrous oxide
NO_2^-	Nitrite
NO_2	Nitrogen dioxide
NO_3^-	Nitrate
NO_y	Oxidised nitrogen (NO, NO_2, NO_3^-, HNO_3, ...)
S	Sulphur
SO_2	Sulphur dioxide
SO_4^{2-}	Sulphate
SO_x	SO_2 and SO_4^{2-}

1 Introduction

In the eighties and nineties, the scientific community has expressed its concern over the long-term effects of nitrogen (N) (e.g. Aber et al. 1989, Nihlgard 1985, Sullivan et al. 1997) and acidifying (e.g. Falkengren-Grerup 1986, Ulrich 1986, Renberg et al. 1993) deposition on forests, grasslands, lakes and streams. Elevated inputs of nitrogen and acidifying deposition have affected natural cycles of many elements, leading to the disruption of numerous natural processes. This introductory chapter aims to present facts and figures of past and present nitrogen and potentially acidifying deposition and their regional distribution in Flanders. Futhermore calculated critical load values for nitrogen and acidifying depositon are discussed and compared to the actual deposition values. Background information is given on pathways of nitrogen and potentially acids in forest ecosystems. We discuss the nitrogen cycle in forest ecosystems and the concepts and consequences of nitrogen saturation and soil acidification. Finally, the aims of this thesis are presented together with a schematic overview of its structure and the relationship between the different chapters.

1.1 Facts and figures for Flanders

1.1.1 Nitrogen deposition

Nitrogen emissions to the atmosphere due to human activities remain elevated in industrialized regions of the world and are accelerating in many developing regions (Galloway et al. 2004). Nitrogen emissions enclose emissions of NO_y, which mainly originates from burning of fossil fuels (the oxidized nitrogen compounds NO, NO_2, HNO_2, HNO_3 and NO^-_3) and of NH_x (the reduced nitrogen compounds NH_3 and NH_4^+) which is mainly emitted by agricultural practices.

Average nitrogen and acidifying depositions in Flanders are still among the highest in Europe (UNECE-EC 2003) and in spite of important decreases in nitrogen deposition (-18% in 2004 in comparison with 1990), the medium-term target for 2010 (adapted from the European National Emission maxima (NEM)) of 29.6 kg $N.ha^{-1}.yr^{-1}$ is still exceeded (Overloop et al.

2006). The contribution of ammonium (NH_4^+) to inorganic nitrogen deposition decreased from 62% in 1990 to 55% in 2004. In 2004 agricultural practices, particularly intensive livestock husbandry, take account for 29% of the total N deposition and about 50% of total NH_4^+ deposition.

Import of nitrogen pollutants from abroad accounts for 50% of the N deposition (Fig. 1.1). So to reduce N deposition, both national and foreign emissions should be proportionally decreased (Overloop et al. 2006).

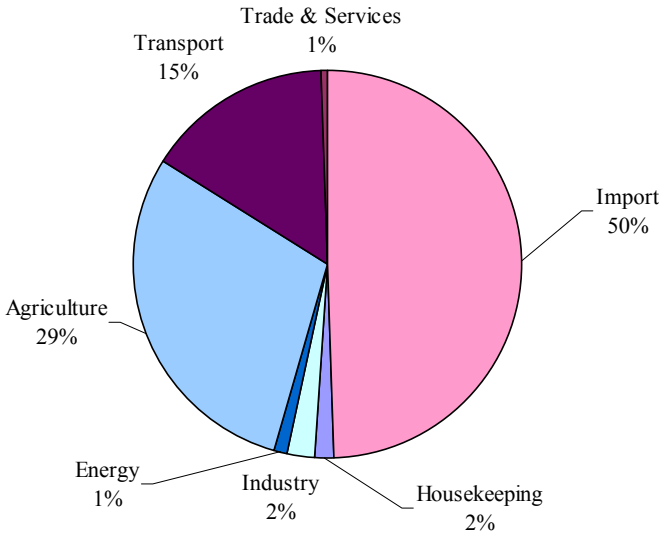

Fig. 1.1: Contribution of different sectors to N deposition in Flanders (source: Overloop et al. 2006)

N deposition is unequally spread in Flanders because of the effect of local emission sources (Fig. 1.2 and Fig. 1.3). NO_y and NH_x have a different behaviour in the atmosphere (Erisman and Heij 1991) and model calculations (Asman et al. 2001) indicate that NH_3 is mainly removed from the atmosphere by dry deposition at a distance less than 1 km from the source or by wet deposition (NH_4^+) at a distance larger than hundred km from the source. NO_x compounds are transported over much longer distances (Erisman and Heij 1991).

Figures 1.2 and 1.3 show the distribution of NH_x and NO_y deposition in Flanders, calculated by means of the atmospheric dispersion model OPS, developed in the Netherlands (Van Jaarsveld 1989) and implemented for Flanders by Mensink & Janssen (1996). This model calculates the transport, the distribution and the deposition of NH_x, NO_y and SO_x based on a resolution of 1x1 km². Input parameters are (1) climatic data (wind direction and velocity,

global radiation, temperature, precipitation duration and amount), (2) land use data and roughness length of the receptors and (3) information of emission sources.

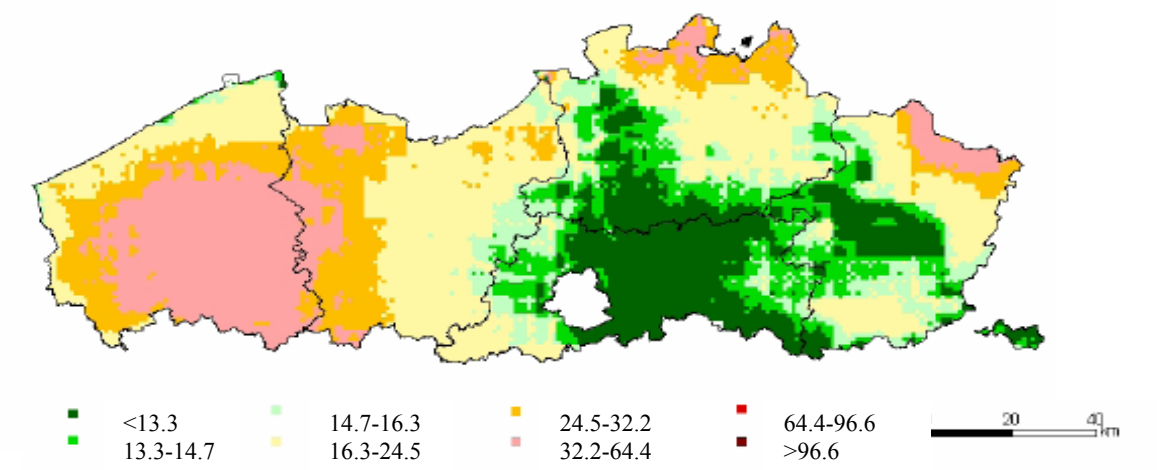

Fig. 1.2: Distribution of NH_x deposition ($kg.ha^{-1}.yr^{-1}$) in Flanders in 2004 calculated by means of the OPS model (Source: Overloop et al. 2006).

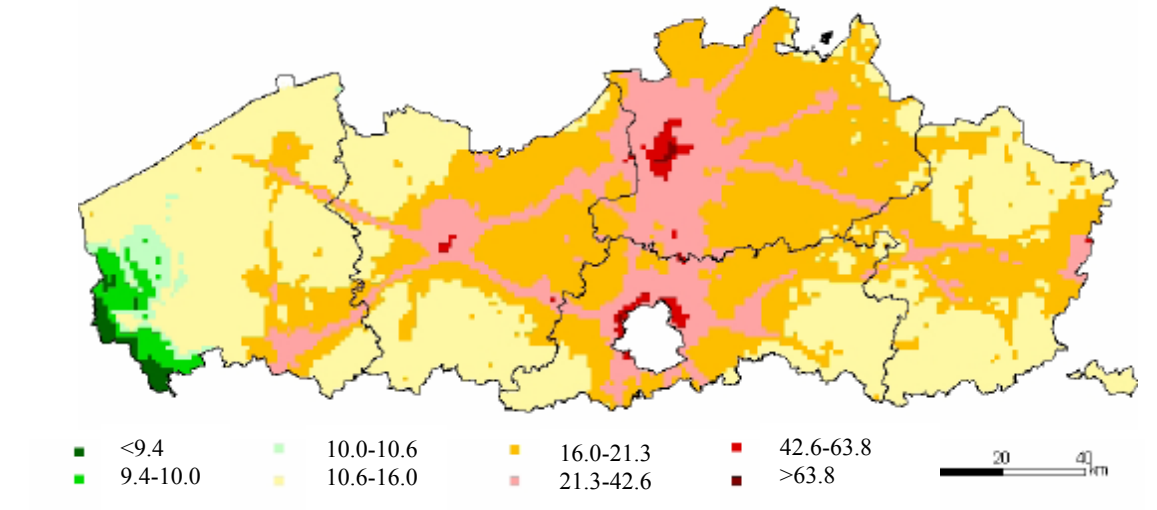

Fig. 1.3: Distribution of NO_y deposition ($kg.ha^{-1}.yr^{-1}$) in Flanders in 2004 calculated by means of the OPS model (Source: Overloop et al. 2006).

The average N deposition varied in 2004 from 22 to 72 kg $N.ha^{-1}.yr^{-1}$ (Overloop et al. 2006). In the centre of the province of West Flanders and in the north-eastern part of Flanders, where intensive livestock husbandry is concentrated, and along main roads, deposition rises above 60 kg $N.ha^{-1}.yr^{-1}$ (Overloop et al. 2006).

On the long-term (2030), the Environment and Nature Plan for Flanders aims to push back the atmospheric nitrogen deposition below the critical load value for (semi-)natural ecosystems.

The concept of critical load (CL) was defined to express the tolerance of natural and semi-natural habitats for anthropogenic air pollution (Nillson and Grennfelt 1988). It relates to the maximum exposure to one or more pollutants without occurrence of any significant harmful effect on for example biodiversity, forest vitality or nitrate seepage to groundwater, according to present knowledge. For Flemish forest ecosystems, critical load values were calculated for 1425 forest plots by Langouche et al. (2002) (Table 1.1) for two criteria: protection of herbal biodiversity and protection of undeep groundwater for nitrate pollution.

The calculations of Langouche et al. (2002) are based on the Statistic Mass Balance method (SMB) (Nillson and Grennfelt 1988). This method is the most commonly used model in Europe for calculation of the critical loads for nitrogen and potentially acidifying depositions for forest ecosystems. The SMB model is based on balancing the nitrogen or potentially acidifying inputs to and outputs from an ecosystem, to derive a critical load value (CL) that ensures a critical chemical limit (related to effects on the ecosystem) is not exceeded. Equation 1.1 allows calculating the critical load value for nitrogen based on knowledge of the soil nitrogen immobilization (N_{im}), uptake of nitrogen by the vegetation (N_{up}) and loss of nitrogen through denitrification reactions (N_{de}). For seepage of nitrogen (N_{seep}), a critical protection criterion is formulated.

$$CL(N) = N_{im} + N_{up} + N_{de} + N_{seep} \quad (1.1)$$

With CL (N) = critical load value for nitrogen, N_{im}=nitrogen immobilization in the soil, N_{up} = nitrogen uptake by the vegetation (and removed when harvested), N_{de}=loss of nitrogen through denitrification reactions and N_{seep}=the allowed nitrate seepage according to the studied criteria.

For protection of groundwater, two NO_3^- concentrations levels in seepage water were used (1) the ecological value of 25 ppm and (2) the drinking water value of 50 ppm. These criteria are each multiplied by the water seepage flux to obtain a critical nitrate seepage flux. The critical load values above which changes in herbal biodiversity occur are based on an available nitrate seepage flux of 1.4 kg N ha^{-1} yr^{-1}. For more information and extensive explanation of the methods we refer to Langouche et al. (2002).

For different vegetation types, these calculated critical load values were compared with the actual level of nitrogen deposition (as modelled by the OPS model) (Overloop et al. 2006). 100% of the area covered by forest ecosystems was exposed to nitrogen deposition levels that are of risk for maintaining their biodiversity (Table 1.1, Fig. 1.4). Even if the medium-term

target of 30 kg $N.ha^{-1}.yr^{-1}$ is achieved in 2010, critical load values will still be exceeded in all forest (with on average 17 kg $N.ha^{-1}.yr^{-1}$) and heathland (with on average 8 kg $N.ha^{-1}.yr^{-1}$) ecosystems (Overloop et al. 2006). For protection of undeep groundwater for nitrate pollution (based on the 50 ppm criterion), the exceedance values are less dramatic: critical load values on the 50 ppm target were exceeded at 12% of the deciduous forest surface area and 69% of the coniferous forest area.

Table 1.1: Median critical load values for nitrogen and their exceedances in forest ecosystems expressed in kg $N.ha^{-1}.yr^{-1}$. For the protection of undeep groundwater, both the values for the 25ppm and 50ppm (between brackets) criteria are given (adopted from Langouche et al. 2002)

	Protection of biodiversity		**Protection undeep groundwater**	
	Coniferous	Deciduous	Coniferous	Deciduous
Median critical load value ($kg.ha^{-1}.yr^{-1}$)	10	15	26 (41)	40 (69)
Median exceedance ($kg.ha^{-1}.yr^{-1}$)*	36	26	22 (7)	0.2 (-27)
% of the forest surface area were the critical load values are exceeded	100	100	99 (69)	53 (12)

* Median exceedance is the median value of the differences between the nitrogen deposition and the calculated critical load value for a total of 1425 forest plots

The lower critical load values, thus the higher sensitivity to nitrogen deposition, for coniferous forest than deciduous forests is caused by (Langouche et al. 2002):

- a lower nitrogen uptake (and removal when harvested) in coniferous tree biomass (especially in stands of *Pinus sylvestris*)
- the frequent occurrence of coniferous forest on well drained soils with low capacity for denitrification (so less N is converted into the harmless N_2 and the greenhouse gas N_2O).
- the lower water seepage flux under coniferous stands (because of higher interception evaporation rates in the tree crowns). Lower water seepage flux calculated with the allowed concentration in the soil solution result in a lower tolerated nitrogen seepage flux.

In the Netherlands (Bobbink and Roelofs 1995; De Vries et al. 1995), critical load values were also calculated for other effects than maintaining biodiversity and protection of groundwater (Table 1.2).

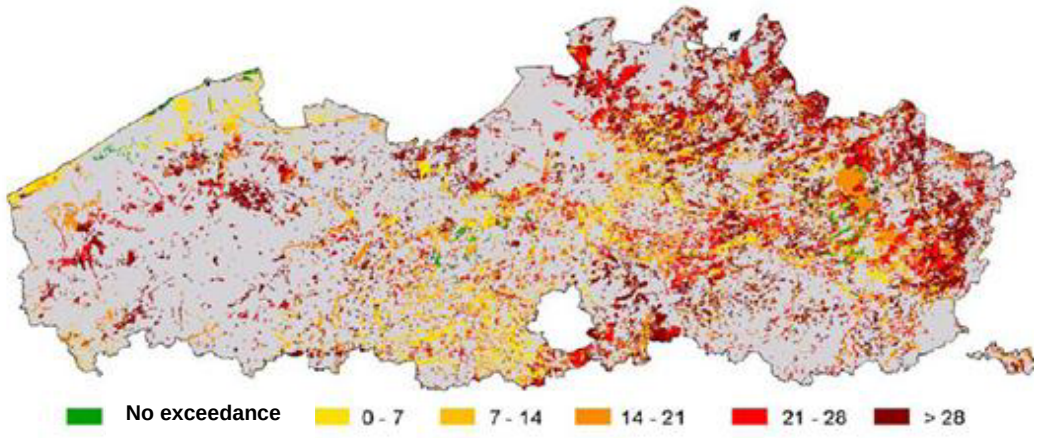

Fig. 1.4: Exceedance (kg $N.ha^{-1}.yr^{-1}$) of the critical load for protecting biodiversity in Flemish forest, heathland and grassland ecosystems in 2004 (Source: Overloop et al. 2006)

Table 1.2: Critical load values ($kg.ha^{-1}.yr^{-1}$) for nitrogen deposition in forest ecosystems and their effects when these values are exceeded (Sources: Bobbink and Roelofs 1995; De Vries et al. 1995). The left-hand values are long-term critical loads, while the right-hand values are calculated for the medium-term.

Vegetation type	**Critical load**	**Effect when exceeded**
Coniferous forest	7-20	Decline of terrestrial lichens and ectomycorrhizae and increase of nitrophilic species
	12.6-21	Nitrate pollution of groundwater
	21-42	Increased tree sensitivity for frost and infestations
	17.5-70	Disturbed nutrient uptake
Deciduous forest	11-20	Decline of terrestrial lichens and ectomycorrhizae and increase of nitrophilic species
	23.5-40.6	Nitrate pollution of groundwater

1.1.2 Potentially acidifying deposition

Potentially acidifying deposition comprises deposition of SO_x, NO_y and NH_x compounds, of which SO_x is the collective term for SO_2 and SO_4^{2-}, NO_y for oxidized nitrogen compounds such as NO, NO_2, HNO_2, NHO_3 and NO_3^- and NH_x for the reduced nitrogen compounds NH_3 and NH_4^+. A first definition of potentially acidifying deposition comprises the summation of SO_x, NO_y and NH_x (UBA 2004). A more complete definition reckons with the neutralizing effect of deposition of so-called base cations (Na^+, K^+, Ca^{2+}, Mg^{2+}), and can include a seasalt correction (for more information we refer to Staelens et al. (2006) and UBA (2004)).

The acidifying effect of the potentially acidifying pollutants NO_3^- and SO_4^{2-} is largely dependent on their chemical composition when deposited. When deposited as a salt (e.g. KNO_3 or $CaSO_4$), no net proton production will occur in the soil (Binkley and Richter 1987), while deposited as an acid (HNO_3 and H_2SO_4), acidification occurs when nitrate or sulphate are seeped from the system with an accompanying base cation. Ammonium (NH_4^+) is only net acidifying when NH_4^+ is nitrified to nitrate (NO_3^-), and two protons are generated (the gas NH_3 acts in the atmosphere or on the plant surface as a base, so consumes one proton to form NH_4^+). Locally however, other possible pathways of NH_4^+ can contribute to acidification of the upper soil horizons. When deposited as NH_4^+, it can be taken up by plant roots ($+1H^+$), assimilated into organic matter ($+1H^+$) and fixed at the cation exchange complex (CEC) ($1H^+$). All these reactions are generating protons and mainly occur in the upper soil horizons, since NH_4^+ is not mobile at all.

The emissions of sulphur (S) have been reduced over much of Europe and the United States by enforced environmental protection policies during the past 25 years (UN-ECE 2003). In Europe, emissions of sulphur dioxide (SO_2) have declined on average by 60% between 1990 and 2000. During the same period, average potentially acidifying deposition in Flanders decreased with about 33% (from 5844 $eq.ha^{-1}.yr^{-1}$ in 1990 to 3925 $eq.ha^{-1}.yr^{-1}$ in 2004) (Fig. 1.5). This decrease was mainly attributable to an average decrease in SO_x deposition of more than 50% and of NH_x deposition of 27% (Van Avermaet et al. 2006).

Similarly to nitrogen deposition (Fig. 1.1), emissions from abroad cause about 50% of the potentially acidifying deposition in Flanders. Agricultural practices (23%) and traffic (12%) are two main acid producers for Flanders (Van Avermaet et al. 2006). Also for nitrogen, sulphur is not equally spread over Flanders. The industrialized region surrounding the harbour of Antwerp receives the highest sulphur deposition (Fig. 1.6).

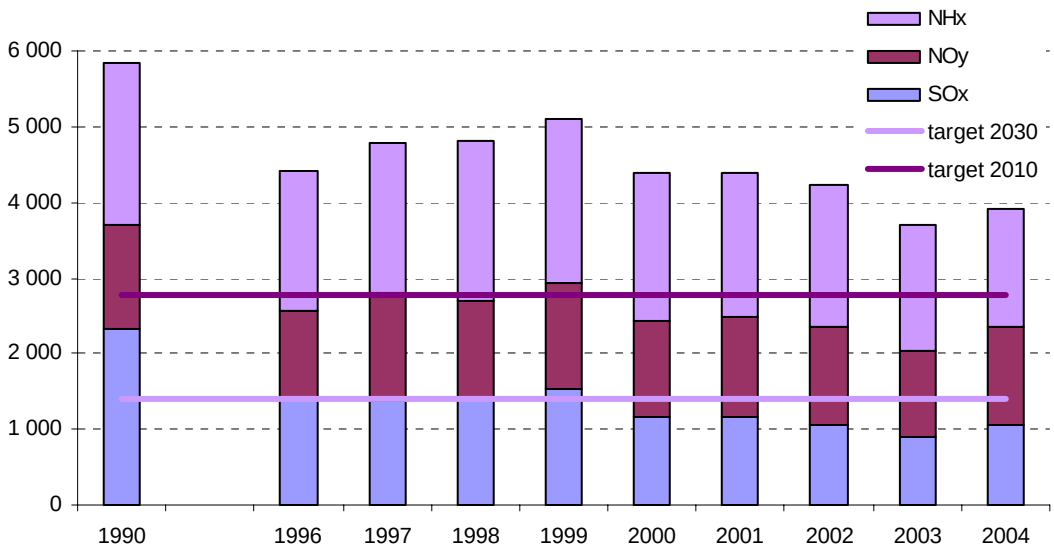

Fig. 1.5: Evolution of potentially acidifying deposition between 1990 and 2004 in Flanders (Source: Van Avermaet et al. 2006)

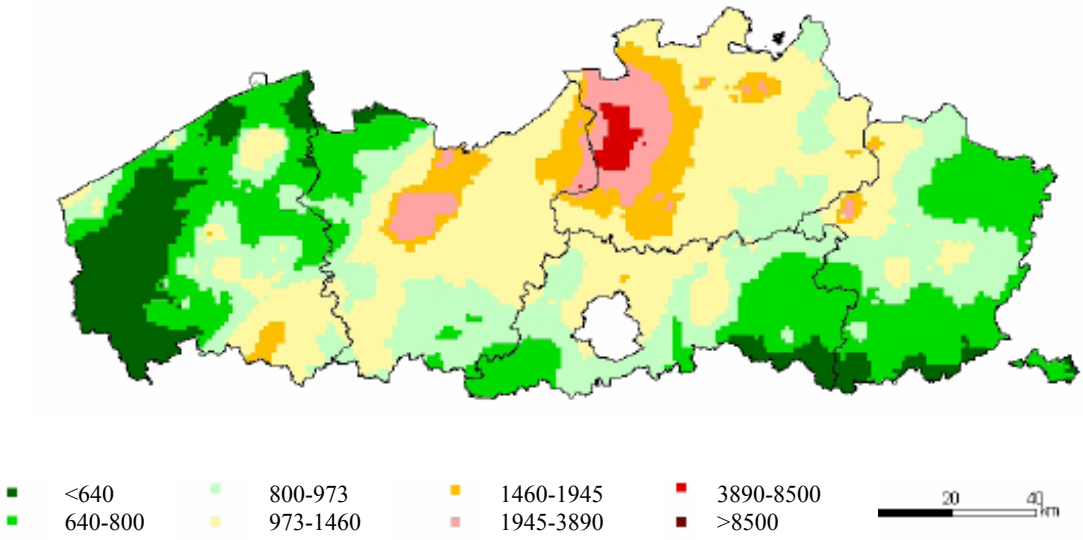

Fig. 1.6: Distribution of SO_4^{2-} deposition ($keq.ha^{-1}.yr^{-1}$) in Flanders in 2004 calculated by means of the OPS model (Van Avermaet et al. 2006)

On the medium-term (2010), potentially acidifying deposition should decrease under the target value of 2770 $eq.ha^{-1}.yr^{-1}$. On the long-term (2030), the Environment and Nature Plan for Flanders aims to push back the atmospheric potentially acidifying depositions under the critical load for (semi-)natural ecosystems. These were calculated for two criteria: tree root damage and soil acidification (Table 1.3). These calculations were based on Equation 1.2.

$$CL_{pot\_acid} = BC_w - BC_{up} + N_{im} + N_{up} + N_{de} - ANC_{seep} \quad (1.2)$$

with $CL_{pot\text{-}acid}$=critical load value for potentially acidifying deposition, BC_w=so-called base cations released by weathering reactions-BC_{up}=uptake of so-called base cations by vegetation (and removed when harvested), N_{im}=nitrogen immobilization in the soil, N_{up} = nitrogen uptake by the vegetation (and removed when harvested), N_{de}=loss of nitrogen through denitrification reactions, ANC_{seep}=seepage of the acid neutralizing capacity of the soil

For protection of the vegetation from root damage, a critical ANC_{seep}-value was defined with a critical ratio of $\frac{Al^{3+}}{BC} = 1$. For the maintenance of the acid neutralizing capacity of the soil, the ANC_{seep}-value was defined to allow no vertical transport of acids through the soil profile. For more information and extensive explanation of the methods we refer to Langouche et al. (2002).

Table 1.3: Median critical load values for potentially acidifying deposition and their exceedances in forest ecosystems expressed in $eq.ha^{-1}.yr^{-1}$, calculated for non-calcareous soils (adopted from Langouche et al. 2002).

	Root damage		**Soil acidification**	
	Coniferous	Deciduous	Coniferous	Deciduous
Median critical load value ($eq.ha^{-1}.yr^{-1}$)	2700	3100	1500	1500
Median exceedance ($eq.ha^{-1}.yr^{-1}$)	830	1100	2100	2700
% of the forest surface area were the critical load values is exceeded	86	95	100	100

1.2 Nitrogen and potentially acidifying deposition in forests

After transport and possible transformations, pollutants eventually deposit on a surface by wet, dry and occult deposition. Wet deposition is defined as the process by which atmospheric pollutants are attached to and dissolved in cloud and precipitation droplets and consequently delivered to the earth's surface by rain. Dry deposition is the process where gasses and particles are deposited directly from the atmosphere onto vegetation, soil or other surfaces without the medium of water. Occult deposition is the process where cloud and fog water

droplets are directly intercepted by the earth's surface. In this thesis, abstraction is made of occult deposition, since its contribution is generally considered to be negligible for lowland forests relative to wet and dry deposition (Vermeulen et al. 1997).

The amounts of wet deposition are hardly influenced by the characteristics of the surfaces on which they are deposited. The quantity of dry deposition is, however, strongly determined by the roughness of the deposition surface. Forests possess surface structures that strongly promote dry deposition of nitrogen and sulphur (e.g. Van Breemen et al. 1982, Ivens 1990), and show, depending on canopy height, canopy closure, leaf area index, longevity, form and shape of foliage (e.g. Augusto et al. 2002), a strong variability in their capacity for capturing dry deposition.

1.2.1 Forests and nitrogen deposition

In pre-industrial times, N has been a major growth limiting factor in (non-agricultural) terrestrial ecosystems, and atmospheric input of reactive N compounds amounted to less than 5 kg $N.ha^{-1}.yr^{-1}$ (Boxman et al. 1998). As a consequence, microbial processes in forest soils, metabolic processes in forest trees, and forest ecosystem functioning are considered to be adapted to N limitation rather than N excess. It is commonly believed that most forests are N limited, and the response to additional N through atmospheric deposition would result in a positive growth response (Tamm 1991).

The artificial inputs of nitrogen highly exceed the natural background values in European forests (MacDonald et al. 2002). Consequently during the past decade, a number of forest ecosystems in Europe and the US have been found to display symptoms of N saturation (e.g. Fenn et al. 1998, Gundersen et al. 2006). Nihlgard (1985) was the first to propose that excessive N deposition could damage forest ecosystems. Responses to N deposition in forest ecosystems are not linear and are therefore not easily captured in simple dose-response functions. Rather, it is expected that these relationships are highly nonlinear, with critical threshold points (Aber et al. 1998).

The N cycle

The N cycle in pristine temperate forests is characterized by an almost closed internal cycle between the primary producers and the large pool of N in soil organic matter (SOM), where the most important processes are litter production (mostly foliage and roots), decomposition, mineralization (including nitrification), immobilization, and plant uptake (Gundersen et al. 2006). Most N in temperate forest ecosystems is bound relatively inactive in soil organic matter (1000-18000 $kg.ha^{-1}$). Only an amount of 200 to 1000 $kg.ha^{-1}$ (or 2 to 20%) is found in plants (trees and ground vegetation) (Gundersen et al. 2006). Both atmospheric and pedospheric sources of nitrogen are available to plants. The contribution of atmospheric N is considered to be low in remote environments and the N demand of plants is met almost exclusively by the uptake of NH_4^+ and/or NO_3^- (Glass and Siddiqu 1995, in Rennenberg et al. 1998) originating from mineralization processes in the soil. Decomposition and mineralization are key processes limiting plant uptake in undisturbed forests, where organic matter tends to accumulate in the soil. Plants may, however, cover part of their nitrogen requirements by uptake of amino acids directly from the organic pool as a result of mycorrhizal symbiosis (Persson et al. 2003). Competition for NH_4^+ and NO_3^- in the soil between plant roots and microbial immobilization, nitrification and denitrification determines N nutrition in plants (Fig. 1.7).

Atmospheric pollution from agriculture (Fangmeier et al. 1994) and fossil fuel burning (Logan 1983) is known to interact with this competition in several ways. Uptake of dissolved NH_4^+ and NO_3^- and gaseous NH_3 and NO_x (Harrison et al. 2000) by aboveground parts of plants can contribute significantly to N nutrition. The input of NH_4^+ and NO_3^- via the precipitation flux alters the physical, chemical and biological properties of the pedosphere and might affect N uptake by roots of plants (Marschner et al. 1991) and lead to nitrogen saturation (see further).

Nitrogen can leave the forest ecosystem through different pathways: (1) release of organic bound N due to forest fire, (2) removal of organic bound N in harvested wood logs in managed forests, (3) seepage of dissolved nitrogen (NO_3^-, NH_4^+ and dissolved organic nitrogen (DON)) via seepage water and (4) gaseous N loss through denitrification reactions in the case of N_2, NO and N_2O and nitrification in the case of NO and N_2O. In pristine forests, seepage of nitrogen generally amounts to less than a few kilograms per hectare per year, and occurs almost entirely in organic form (DON) (Campbell et al. 2000, Hedin et al. 1995, Perakis and Hedin 2002).

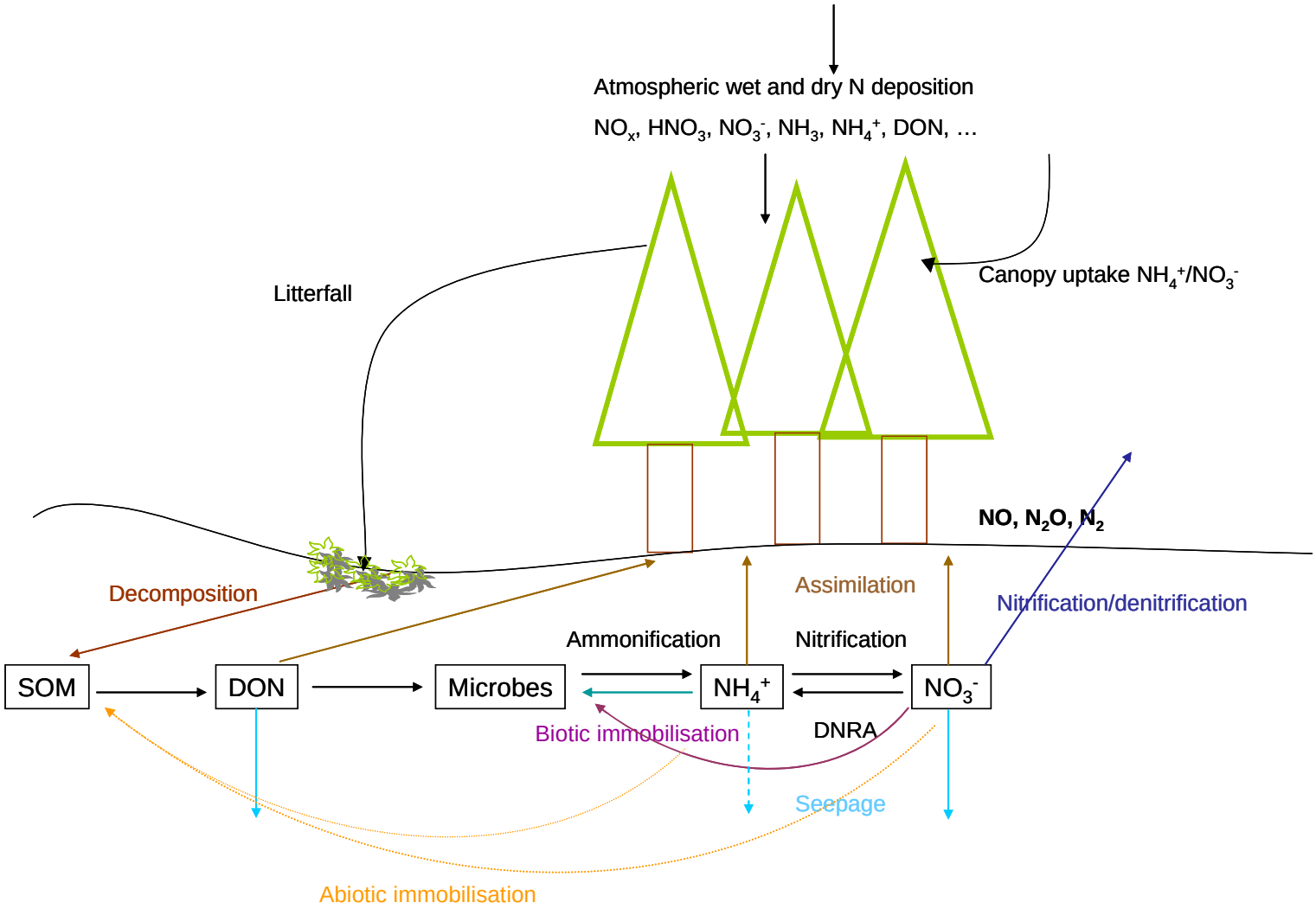

Fig. 1.7: Simplified model of the N cycle in forests (partly adopted from Rennenberg et al. 1998, Davidson et al. 2003 and Gundersen et al. 2006, Schimel et al. 2004).

What is a nitrogen saturated ecosystem?

N input from air pollution may lead to a disruption of the tight retention of plant available N in forests and lead to nitrogen saturation. Various definitions of nitrogen saturation have been proposed:

- an ecosystem unable to absorb additional N (Agren 1983);
- an ecosystem where the primary production will not be further increased by an increase in the supply of N (Nilsson 1986);
- an ecosystem where N losses approximate or exceed the input of N (Agren and Bossata 1988);
- an ecosystem where the availability of ammonium and nitrate is in excess of total combined plant and microbial nutritional demand (Aber et al. 1989).

All these definitions can be understood as stages of nitrogen saturation related to different components in the ecosystem. A forest ecosystem can, for example, leach nitrate, so being nitrogen saturated according to the first definition, but still respond to N additions (e.g. in the spring) and accumulate a considerable amount of nitrogen in the biomass.

In general, the term 'nitrogen saturation' should be seen as a conceptual model predicting that a N-limited system initially retains anthropogenic N by using it for plant and microbial growth as well as for accumulation in biomass and soil organic matter. At some point, however, N inputs begin to exceed the abiotic and biotic N demands within the ecosystem. The system is then predicted to lose its nitrogen retention capacity. As this capacity is exceeded, excess N is available to be lost from the system via seepage water and gas fluxes. Nitrate seepage has been identified as a primary sign of N saturation in N-limited temperate forests if it occurs

- at a significantly higher rate than background levels (5 kg $N.ha^{-1}.yr^{-1}$, Boxman et al. 1998);
- aseasonally, so during the complete year, and not only during the dormant season (Matson et al. 2002).

Overall, N saturation implies a permanent change in the functioning of the N cycle from a virtually closed internal cycle to an open cycle where excess N is leached and/or emitted from the system.

The development of N saturation by increased N inputs involves a complex interaction of the processes in the N cycle (Aber et al. 1998). A simple schematic illustration of the progression of a forest ecosystem from N limitation to N excess and the potential effects of N deposition has been given by Gundersen et al. (2006) and presented in Fig. 1.8. In the first phase, primary production increases. Plants and microbes effectively absorb added N and the N content of the plants increases. In response to this increase in N availability the shoot/root ratio, leaf area and leaf N concentration might all increase, leading to a higher level of net photosynthesis and, consequently, to increased tree growth. Retranslocation of N from senescent foliage (and roots) may decrease, leading to higher N contents in litter materials and thus increased litterfall N flux. More N would then be returned to the soil via above-ground and below-ground litter and, as a result, more N would be mineralised in connection with litter decomposition, which would further enhance N availability.

The internal cycling of N is thus accelerated through different causes: increases in N litterfall, net mineralization and tree N uptake. As N availability is increased, the composition of the forest floor vegetation may gradually change towards more nitrophilic species and other essential nutrients (P, K, Ca, and Mg) or water may at least periodically limit tree growth. The ecosystem approaches N saturation when elevated nitrate seepage becomes a chronic condition and consequently soil acidification from N transformation becomes significant. After reaching these N saturated conditions, N seepage will continue to increase with deposition and forest growth might decline. Recent synthesis efforts support this general scheme although the understanding of processes and interactions is very complex, as has been described in more detail by e.g. Aber et al. (1998), Emmett et al. (1998a, 1998b), Fenn et al. (1998), and Gundersen et al. (1998b, 2006).

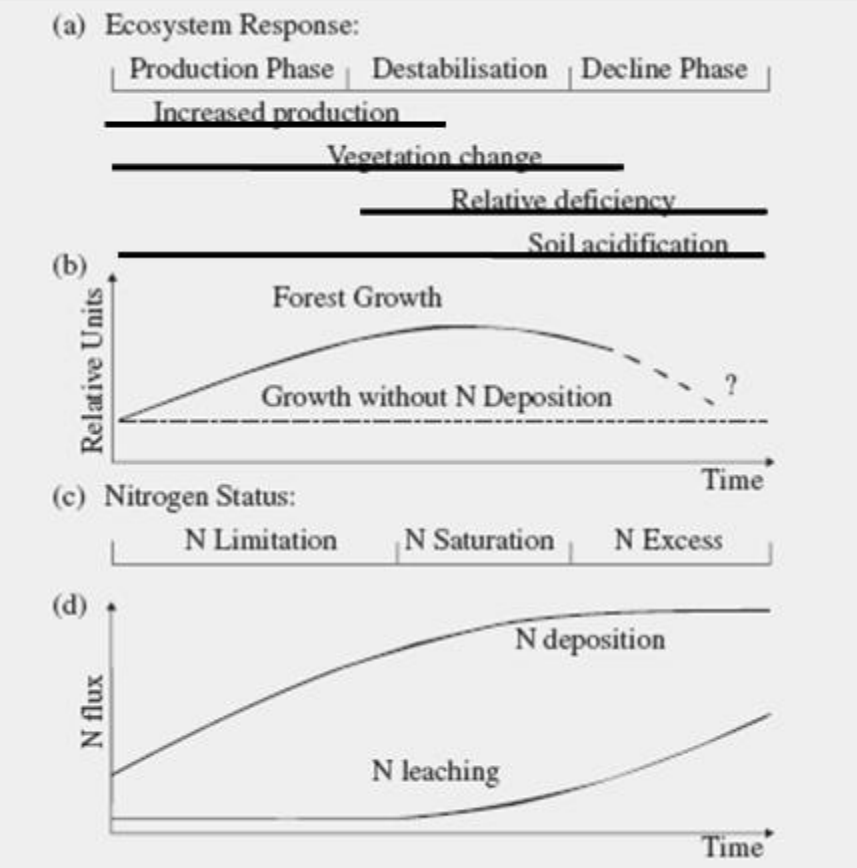

Fig. 1.8: Hypothesized responses of managed temperate forest ecosystems to increased N deposition (a) ecosystem responses (relative deficiency refers to water and other nutrients than N), (b) relative changes in growth, (c) N status and (d) input-output relations. The time scale (x axis) for the response may differ widely between ecosystems and regions (adopted from Gundersen et al. 2006)

Consequences of nitrogen saturation

Nitrification is certainly an ecological key process, since it converts less readily mobile ammonium into mobile nitrate that is easily leached out of the soil profile. Furthermore, the process produces protons that acidify the soil according to the reactions (Equation 1.3):

$$NH_4^+ + \frac{3}{2}O_2 \longrightarrow NO_2^- + 2H^+ + H_2O$$
$$NO_2^- + \frac{1}{2}O_2 \longrightarrow NO_3^- \qquad (1.3)$$

Besides nitrate seepage, potential soil effects of N saturation therefore also include increased soil acidification (Van Breemen et al. 1982), aluminium mobility (Johnson et al. 1992a) (see §1.2.2), elevated so-called base cation seepage (Fenn et al. 1998) (see §1.2.2) and altered

emissions and consumption of greenhouse gases (N_2O, NO, CH_4) (Steudler et al. 1989, Castro et al. 1994, Butterbach-Bahl et al. 2002, Ventera et al. 2003).

The atmospheric concentration of nitrous oxide (N_2O) has increased in recent decades (Kesik et al. 2005) due to anthropogenic perturbation of the global N cycle (Galloway et al. 2004) and hence the elevated rates of atmospheric N deposition to many forest ecosystems in Europe, North America and Asia. Among other sources, forest soils have been acknowledged to represent significant sources of this potential greenhouse gas (Skiba et al. 1994), and several publications evidence that N depositions to forest ecosystems are positively correlated with N_2O emissions (Brumme and Beese 1992, Butterbach-Bahl et al. 1998).

Forest soils have also been shown to act as primarily sources of NO, a reactive trace gas involved in the production of ozone in the troposphere (Butterbach-Bahl et al. 2002). However, due to its reactivity, only a part of the NO emitted from forest soils will reach the atmosphere, whereas some of the NO will react with ozone to NO_2, associated with a partial deposition of NO_2 to plant and soil surfaces or an uptake by plant tissues (Dorsey et al. 2004). As for N_2O, atmospheric N deposition to forest ecosystems has been shown to be closely related to emissions from the forest soils of NO (Pilegaard et al. 1999). The emissions of NO and N_2O in relationship to the forest type are briefly discussed in $ 8.6.

Will forest growth decline at conditions of N excess?

Several authors report an increase in forest growth as a consequence of high atmospheric N input (Spiecker 1990, Hunter and Schuck 2002, Solberg et al. 2004). Long-term continuous N inputs may, however, lead to decreases in net primary production (NPP) (especially in wood growth; Aber et al. 1998, Emmett 1999) and, in some cases, to forest decline (McNulty et al. 1996). Although many studies applied large fertilizer doses that exceed realistic rates of N deposition, a few have documented NPP decline in response to very low levels of N additions. For example, decreased growth of both coniferous and hardwood species has been observed after six years of N additions in the range of 16 to 31 kg N ha^{-1} yr^{-1} in experimentally fertilized, high elevation spruce-fir stands in the north-eastern US (McNulty et al. 1996). Nevertheless, significant positive forest growth was observed during the first three years. In European case-studies, NPP was increased by 50% after ambient levels of atmospheric N deposition were experimentally decreased using rain-out shelters with clean water (Boxman et al. 1998, 2007). Doherenbusch et al. (2002) did, however, not find an increase of growth rate

and aboveground biomass after clean rain treatment from 1991 and 1998, although increases in fine root biomass were reported for the same plot (Lamersdorf et al. 2004).

Nitrogen cycling and climate change

Climate change induces changes in temperature, precipitation patterns and atmospheric CO_2 concentrations. These factors may directly or indirectly influence decomposition and mineralization of organic matter, which are key-processes in the N cycle of a forest ecosystem. Few experimental data however are available from forest ecosystems and in most cases only one factor and or one part of the ecosystem have been manipulated.

Rustad et al. (2001) published a meta-analysis of 12 warming experiments, among which 3 forest sites, showing an overall response of increased but not significant net N mineralization. A deciduous stand with complete N retention at Harvard Forest (Melillo et al. 2004) showed increased net N mineralization as a reaction to soil warming. All nitrogen was retained within the ecosystem and no effect on nitrogen seepage fluxes was noted. In a nitrogen saturated pine forest in Norway however (Lükewille and Wright 1997), soil warming increased both net N mineralization and nitrate seepage. In experiments on heathlands across Europe, Schmidt et al. (2004) found a significant increase in nitrate seepage in response to soil warming.

Norby et al. (1999, 2001) established field-scale experiments with increased atmospheric CO_2 and found changes in litter quality during the first years of treatments: decreased N concentration and increased lignin concentration due to the direct positive effect on photosynthetic activity. No consistent influence on decomposition could however been shown. Another study reported by Finzi et al. (2001), Oren et al. (2001) and Schlesinger and Lichter (2001) shows increased storage of C in the upper soil layers and biomass, but fast turnover rates of organic carbon in the litter layer appear to constrain the potential size of this carbon sink. Given the observation that carbon accumulation in the deeper mineral soil layers was absent, the authors suggest that significant long-term net carbon sequestration in forest soils is unlikely. This study could not detect changes in litter N concentration or soil nitrate and ammonium concentrations. No data on seepage water nitrate have been reported, but in general Gundersen et al. (2006) expects an increased nitrogen retention in response to CO_2 enrichment alone through the tree growth response. So changes in nitrate seepage in response to climate change will depend on the ability of plants to balance the increased nitrogen mineralization by additional N uptake by the vegetation and on the N-status of the system.

According to Gundersen et al. (2006), it is likely that N seepage will increase from N saturated systems, whereas the effect of warming and CO_2 will increase N demand in more N-limited systems.

The role of forests in the 'Nitrogen Cascade'

Galloway et al. (2003) defined the phenomenon of 'the nitrogen cascade' as being the sequential transfer of reactive nitrogen through environmental systems, resulting in environmental changes as nitrogen moves through or is temporarily stored within each system (Fig. 1.9). For an extensive explanation of this concept, we refer to Galloway et al. (2003), but the role of forests within this phenomenon will be briefly discussed here.

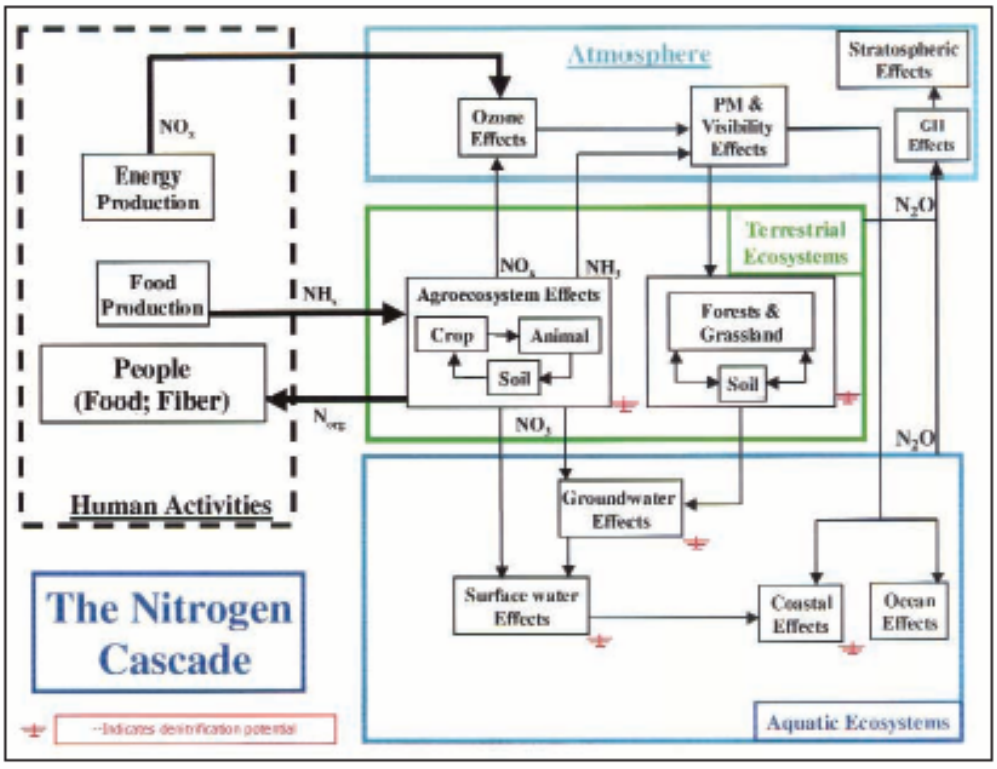

Fig. 1.9: Illustration of the nitrogen (N) cascade concept introduced by Galloway et al. (2003), showing the sequential effects that a single atom of N can have in various reservoirs after it has been converted from a nonreactive to a reactive form. Abbreviations: GH=greenhouse effect, NH_3= ammonia, NO_3^-=nitrate, NO_x=nitrogen oxide, N_2 =nitrous oxide, PM=particulate matter. Adopted from Galloway et al. (2003).

Forests can be a major reservoir and a short- to long-term sink within the N cascade. In unpolluted regions, inputs of reactive nitrogen can range between 1 and 2 kg $N.ha^{-1}.yr^{-1}$ while in polluted regions it can reach values up to 100 kg $N.ha^{-1}.yr^{-1}$. Outputs as dissolved organic N (DON) are low and relatively constant across sites (e.g. Goodale et al. 2000), while dissolved inorganic N (DIN) loss is quite variable and can be as low as zero or equal to deposition, depending on forest history, emission regime and tree species composition (Dise et al. 1998, Gundersen et al. 1998, Fenn et al. 2003).

In general, forests can create a significant lag in the N cascade because of the high residence time of reactive nitrogen within a forest (can be hundreds to thousands of years). Human activity can shorten this residence time either by increasing inputs or by harvest practices, fires or conversion to agriculture (Gundersen et al. 2006). Most N is held in forest soils (Nadelhoffer et al. 1999), and a better understanding of the kinetics and capacity of soils is a key in predicting nitrogen retention rates (so the development of N saturation) and the role of forests in the N cascade.

Knowledge gaps

To understand the changes in N cycling when a forest ecosystem evolves from N limited to N saturated, experiments were set-up in Europe and in the United States focusing on (1) the effects of increasing nitrogen input by fertilization (e.g. Carnol 1997, Gundersen et al. 1998b, Magill et al. 1997, 2000, 2004, Aber et al. 2004) and (2) the reduction of nitrogen and potentially acidifying inputs by roof manipulations (e.g. Tietema et al. 1995, Gundersen et al. 1998b). The results from N reduction experiments have shown that nitrate seepage is efficiently reduced to very low levels in a rather short-term (1–2 years after the start of manipulation) (e.g. Bredemeier et al. 1998a, 1998b).

Despite the rapid increase in our understanding of N saturation in forests, two critical aspects of N saturation remain unclear (Aber and Magill 2004). The first is the incomplete knowledge of the processes of soil retention. Even stands in Central Europe which have experienced elevated N inputs for decades retain, on average, about 70% of current inputs (e.g. Dise et al. 1998; MacDonald et al. 2002). As most of the negative effects of N saturation are related to increased N cycling, nitrification and nitrate seepage (Aber et al. 1998), understanding the nature, kinetics and capacity of N retention mechanisms is critical for predicting the regional onset of saturation.

The second gap in knowledge is the capacity of N saturation to reduce forest growth and increase tree mortality. While cases of forest decline and death have been linked either to N deposition or interactions of N with other elements and/or infestations (e.g. Schulze, 1989; McNulty et al. 1991, 1996; Magill et al. 2000, 2004) relatively little is understood about the mechanisms by which damage occurs or about the physiological basis for this response (e.g. DeHayes et al. 1999a, 1999b, Bauer et al. 2004).

1.2.2 Forests and potentially acidifying deposition

Acidification of forest soils in Europe and North America has been an important concern over the last decades. A large number of recent studies report a significant forest soil acidification over the past 50 years (e.g. Ahokas 1997, Drohan ad Sharpe 1997, Dai et al. 1998, Hovmand and Hansen al. 1999, Van der Salm and De Vries 2000, Miller et al. 2001, Heisner et al. 2003). Many of these studies indicate potentially acidifying deposition as one of the major causes.

Definition of and processes causing soil acidification

Soil acidification is the build-up of hydrogen cations, also called protons, in the soil. A soil acidifies when its acid neutralising capacity decreases and the proportion of K^+, Ca^{2+} and Mg^{2+} on the clay-humus-exchange complex decreases in advantage of aluminium and iron (De Vries and Breeuwsma 1985).

Soil acidification occurs by a flux of protons to the soil. Table 1.4 gives an overview of the most important proton producing and consuming processes in terrestrial and aquatic ecosystems.

How does a soil buffer incoming protons?

With respect to the soil matrix, the solid phase accepting protons and releasing cations changes with changing soil pH. This leads to a sequence of successive buffer reactions with decreasing pH (Fig. 1.10 and Table 1.5). The given pH limits have been calculated by Ulrich (1986) and may vary, depending on the composition of the buffer substances and on how much the soil state deviates from chemical equilibrium.

Table 1.4: Proton producing and consuming processes (Adopted from Van Breemen et al 1985)

Proton producing processes	**Proton consuming processes**
Atmospheric input of acid or potentially acid substances	Export of acid or potentially acid substances via drainage water
Net-assimilation of cations by vegetation (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, NH_4^{+})	Net-mineralization of cations from organic matter (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, NH_4^{+})
Net mineralization of anions from organic matter ($H_2PO_4^{2-}$, NO_3^{-}, SO_4^{2-})	Net assimilation of anions by the vegetation ($H_2PO_4^{2-}$, NO_3^{-}, SO_4^{2-})
Deprotonation of weak acids	Protonation of anions
Oxidation reactions	Reduction reactions
Precipitation of cations (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+} Al^{3+})	Weathering of cations (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+} Al^{3+})
Mineral weathering of anions ($H_2PO_4^{2-}$, SO_4^{2-})	Precipitation of anions ($H_2PO_4^{2-}$, SO_4^{2-})

Carbonate and silicate buffer range

In the carbonate and silicate buffer range, the reaction products of protons with $CaCO_3$ are water-soluble bicarbonates which are leached from the soil with the seepage of water. As soon as $CaCO_3$ is depleted, the so-called base cations leached as bicarbonate originate from silicate weathering. The weathering of primary silicates, which takes place under all pH conditions, is the dominant buffer reaction in soils free of carbonates at pH > 5. The products of weathering are clay minerals with permanent negative charge. The cations liberated during silicate weathering can be bound as exchangeable cations in the clay minerals formed from the weathered silicate lattices. The seepage of HCO_3^{-} will approach zero if pH approaches 5 since H_2CO_3 dissociation becomes negligible at this pH.

Cation exchange buffer range

At a soil pH < 5, the so-called base cations of the cation exchange complex of clay minerals and organic matter, mainly Ca^{2+}, play a role in buffering protons. At the same time, the solubility of the oxides of manganese and other metals reach ecological significant concentrations (Hildebrand 1986). Also the rate of aluminium release from the silicate lattices increases. The main reaction products are non-exchangeable polymeric aluminium hydroxo-

compounds and aluminium in ionic form (Al^{3+}). The non-exchangeable polymeric aluminium hydroxo-compounds accumulate in the interlayer of swelling clay minerals, blocking the permanent charges, and so reduce the cation exchange capacity (CEC). At the same time, the Al^{3+} ions can occupy exchange sites, resulting in a reduction of base saturation (which is the percentage of exchange sites occupied by the so-called base cations). Due to the highly selective binding of Al^{3+} on these exchange sites, the Al^{3+} saturation can reach high values at low Al^{3+} concentrations in soil solution.

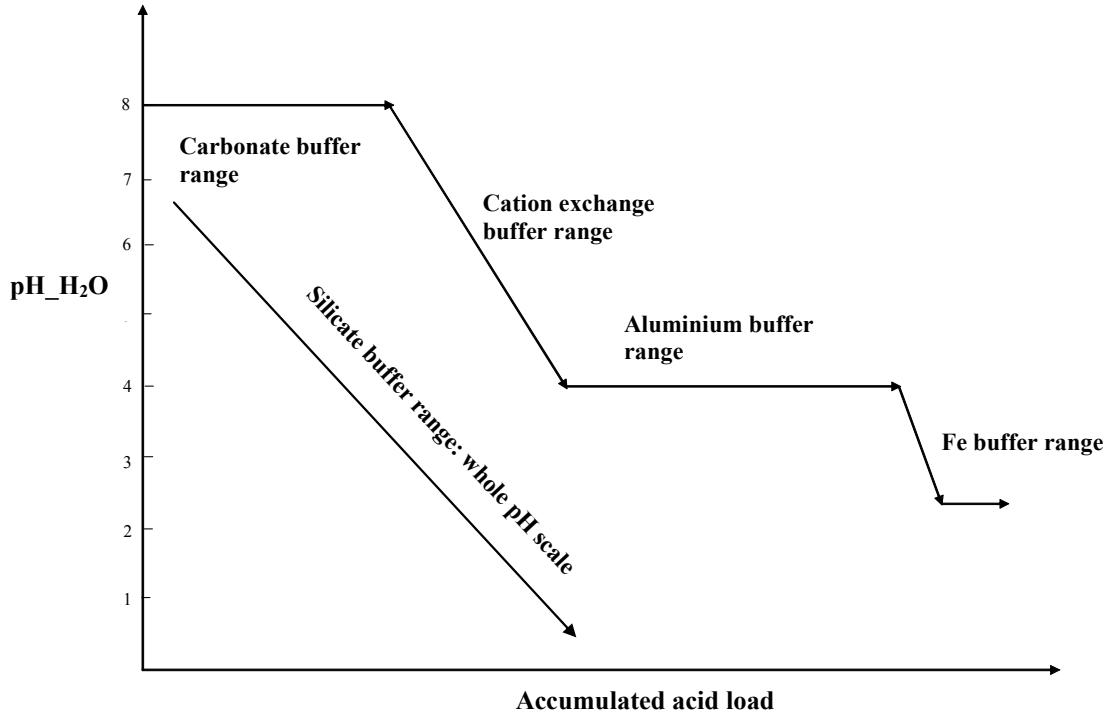

Fig. 1.10: Buffer ranges and changes in pH-H_2O in the course of soil acidification (after Ulrich 1991)

If the acid loading is due to H_2SO_4, the formation of amorphous Al-hydroxy-sulphates is possible (Equation 1.4) and the soil reacts as a sink for H_2SO_4 (Ulrich 1991). This accumulation seems only to be possible at pH values higher than 4.2.

$$Al(OH)_3^s + H_2SO_4 \rightleftharpoons AlOHSO_4^s + 2H_2O \qquad (1.4)$$

Table 1.5: Overview of buffer systems and their pH ranges in the soil (adopted from Ulrich 1991)

Buffer substance	**pH range**	**Main reaction product of lower acid neutralising capacity (ANC)**	**Chemical change in soil**
Carbonate buffer range			
$CaCO_3$	8.6>pH>6.2	$Ca(HCO_3)_2$ in solution	Seepage of Ca^{2+} and HCO_3^-
Silicate buffer range			
Primary silicates	Whole pH scale (dominant in soils pH>5)	Clay minerals	Increase of CEC
Cation exchange buffer range			
Clay minerals	5>pH>4.2	Nonexchangeable $n[Al(OH)_x^{(3-x)+}]$	Blockage of negative permanent charge reduction of CEC
Mn-oxides		Exchangeable Mn^{2+}	Reduction of base saturation
Clay minerals		Exchangeable Al^{3+}	Reduction of base saturation
Interlayer Al $n[Al(OH)_x^{(3-x)+}]$		Al-hydroxo-sulphates	Accumulation of acid in case of input of H2SO4
Aluminium buffer range			
Interlayer Al Al-hydroxo-sulphates	4.2>pH	Al^{3+} in solution	Al displacement, reduction of permanent charge
Aluminium/iron buffer range Like Al buffer Soil $Fe(OH)_3$	3.8>pH	Fe^{2+} and H^+ in solution	Fe^{2+} displacement, bleaching
Iron buffer range			
Ferrihydrite	3.2>pH	Fe^{2+} and H^+ in solution	Fe^{2+} displacement, bleaching, clay destruction

The so-called base cations previously bound at the exchange sites are leached together with the anion of the acid-generating the proton. The base saturation drops from high to low values (<5%) and the CEC is strongly reduced because the permanent charges are blocked by covering of clay surfaces with the Al-hydroxy-compounds.

Aluminium buffer range

At a pH of 4.2, the solubility of Al-hydroxy compounds increases to an extent that Al^{3+} may become the dominating cation in the soil solution and is being leached from the soil with seepage water. Storage of Al-hydroxy-compounds and sulphates which have been accumulated in the exchanger buffer range then decreases. This buffer range certainly shows a tendency for sulphate and acidity output exceeding the input from potentially acidifying deposition. The storage of exchangeable so-called base cations no longer represents a buffer capacity if the soil is in this buffer range.

At pH < 3.8, Fe oxides are included in the buffer reactions in the presence of dissolved organic matter. This is accompanied by a shift in the exchangeable cation composition from Al^{3+} to H^+ and Fe^{2+} ions. In soils low in reactive iron oxides, proton buffering ceases and the H^+ output approaches its input.

Iron buffer range

At pH values < 3.2, in horizons influenced by infiltrating organic matter with a pH below 3.8, the solubility of iron oxides becomes high enough to reach iron concentrations in the soil solution of ecological significance. The transport of iron leads to visible colour symptoms in the soil profile, which is not the case for aluminium.

Can forest soils recover from soil acidification?

In the last 20 years, anthropogenic emissions of SO_2 together with mineral dust and subsequent deposition of H^+, SO_4^{2-}, Ca^{2+} and Mg^{2+} to forest ecosystems in Europe and North America have decreased substantially (Stoddard et al. 1999; Hedin et al. 1994). Based on the emissions in 1980, clean air protocols (e.g. the Göteborg Protocol 2003) led to a reduction of SO_2 emissions by about 60% in Europe and 20-30% in North America in 2000 (UN-ECE,

2003). This indicates that stress by potentially acidifying rain on all types of ecosystems is diminished. While streams and lakes have shown rapid recovery in response to deposition reductions (Fölster and Willander 2002, Forsius et al. 2003, Stoddard et al. 1999), forest soils have responded more slowly or not at all (Martinson et al. 2005, Hogberg et al. 2006, Oulehle et al. 2006).

The previous high rates of potentially acidifying deposition raise questions concerning the time scale of soil and forest ecosystem recovery. Recent papers highlight that the deterioration of the soil environment with respect to potentially acidifying stress and reduced so-called base cation availability has continued (Oulehle et al. 2006). The release of formerly stored SO_4^{2-} from the soil can markedly delay the recovery of soil solution and streams for decades (Mörth et al. 2005; Novák et al. 2000). The mobilization of this inorganic sulphate causes H^+ release into the soil solution and delays the mitigating effects of decreasing sulphate and H^+ inputs on soil pH and aluminium concentration in the soil solution. Soils with a high storage capacity for sulphate and protons indicate that reversing acidification can take decades because the release of previously stored sulphate causes cation seepage and acidification of deeper soil layers and waters (Morth et al. 2005, Novák et al. 2000).

A recent publication of Hogberg et al. (2006) contradicts with the latter paragraph. Ten years after the termination of high amounts of N additions, several significant indications of recovery were noted: the levels of extractable inorganic N decreased, exchangeable so-called base cations increased and soil pH increased. Deacidification indeed can take place if the rate of proton production becomes lower than the rate of proton consumption and release of so-called base cations by means of either silicate weathering or mineralization of organic matter (Ulrich 1991). In highly acidified soils, recovery of soils totally depends on cation release from silicate weathering. According to Ulrich (1991), however, it can take decades to centuries in managed forests to return to a base saturation in the rooting zone exceeding 15% by means of silicate weathering.

The process of silicate weathering is reported to be influenced by the tree species (Augusto et al. 2006). Furthermore, tree species with high Ca contents in their leaves/needles can positively influence base saturation of the forest floor (Reich et al. (2005) (see also Chapter 6). Reich et al. (2005) observed rapid (within three decades) changes in soils beneath different tree species. Species affected soils both directly through the chemistry of their litter and indirectly through the effect of their litter on detrivores, including earthworms (Reich et al. 2005). Litter Ca appeared to be a critical and general agent in these processes. Vegetation, soil

organisms and soil chemistry are co-developing components of terrestrial ecosystems (Muys et al. 1992, Van Breemen 1993, Berendse 1998, Binkley and Giardina 1998 and Ponge et al. 1999) what highlights the need to consider the role of the tree species as 'ecosystem engineer' for ecosystem recovery.

Furthermore, some authors did find a decreasing deposition of the so-called base cations in Europe and North America (Hedin et al. 1994, Meesenburg et al. 1995), what can be reflected in decreasing soil solution concentrations when exchangeable pools of these elements in the soil are small (Alewell et al. 2000). Together with continuous aluminium stress caused by low soil pH, this can lead to worsening Ca^{2+} and Mg^{2+} supplies for the tree assimilatory tissues (Alewell et al. 2000, De Wit et al. 2001). It is very difficult to demonstrate aluminium toxicity of tree species under field conditions (Weber-Blaschke et al. 2002). However, pot experiments with Norway spruce (*Picea abies* L. Karst) seedlings, a tree species that is usually considered to be an acid adapted tree species in the temperate zone (De Wit et al. 2001, Weber-Blaschke et al. 2002, Zhong et al. 2003), revealed that root and needle uptake of Ca^{2+} and Mg^{2+} were reduced when a threshold concentration of 100 µmol $Al^{3+}.l^{-1}$ was exceeded, and morphological damage to root tips occurred at 400 µmol $Al^{3+}.l^{-1}$ (Godbold 1994, Puhe and Ulrich 2001). Also more demanding species as ash (*Fraxinus excelsior* L.) and sycamore (*Acer pseudoplatanus* L.) were shown to suffer from high concentrations of aluminium in the soil solution (100 µmol $Al^{3+}.l^{-1}$) through direct damage of the morphological structures and the function of roots in the case of ash and antagonistically inhibition of Ca^{2+} and Mg^{2+} uptake in the case of sycamore (Weber-Blaschke et al. 2002). Although these experiments deliver interesting results from the physiological and histological point of view, it is however difficult to transfer the findings to older trees and to field conditions.

Furthermore, the aluminium concentration in the soil solution might be a weak indicator for Al toxicity because Al^{3+} and several Al hydroxides (in this work summarized by the term Al(III)) with distinct toxicity generally coexist in the soil solution within the aluminium buffer range (Sparks 1995). As indicators for tree root damage, molar ratios of Mg^{2+}/Al^{3+} (Jorns and Hecht-Buchholz 1985), Ca^{2+}/Al^{3+} (Cronan and Grigal 1995) and $(K^{+}+Ca^{2+}+Mg^{2+})/Al^{3+}$ (Sverdrup and Warfvinge 1993; Hruska et al. 2001) are commonly used. The item of forest recovery is also discussed in § 8.9.

1.3 Aim and schematic overview of the thesis

Over the past decades, a tremendous effort has been made to better understand all aspects of the topics 'forest and nitrogen deposition' (1961 hits in Web of Science (period 1972-2007), http://portal.isiknowledge.com/), 'forest and nitrogen saturation' (658 hits) and 'forest and acidification' (1605 hits). This thesis adds information to the present knowledge for three main reasons:

1. Nitrogen and potentially acidifying depositions in Flanders are among the highest in Europe (UNE-ECE 2003). This allows studying the 'worst case scenario' of soil acidification and element seepage in forest ecosystems. The present thesis focuses on forest ecosystems on sandy soil, which are very vulnerable for soil acidification and often situated in regions with high NH_3 emissions.
2. The nowadays experimental evidence and datasets of element deposition and seepage quantities in (semi-)natural ecosystems are dominated by coniferous forest (Gundersen et al. 2006). Less is known about deciduous forest ecosystems and only limited comparisons are made between different forest types in similar site and climatic conditions (De Schrijver et al. in press). Moreover, comparisons of forest with other vegetation types, e.g. heathland, are rarely existing (e.g. Herrmann et al. 2005).
3. The amount of research that has been performed on the interaction with and impact on these environmental problems of different forest types is surprisingly limited. This topic is, however, of great interest because, besides its fundamental value, this information is useful in the light of setting the daily forest conversion goals, both on a Flemish and a European scale. Forest management in Flanders is 'ready' for the conversion of homogeneous pine plantations into mixed forests based on the species *Pinus sylvestris* L., *Quercus robur* L. and *Betula pendula* L. The main target being more multifunctional forest ecosystems, delivering an optimal spectrum of ecosystem services. At present, the Flemish government considers a conversion towards mixed forests as the optimal approach to sustainably achieve these goals, from the ecological, the economical and recreational point of view.

This thesis therefore aims at providing useful information on the impact of different forest types on the nitrogen and potentially acidifying deposition load to the forest floor and the

consequences of it. Furthermore, this information is relevant in the light of ecosystem recovery under lower deposition loads aimed at the long-term.

Forests of Scots pine are distributed far beyond their assumed natural range in Europe. Currently Scots pine forest has a share of 31% in the total European forest area (thirty countries) (Spiecker *et al* 2004). Also in Belgium large surfaces of homogenous conifer plantations are on sites naturally dominated by broadleaved species. In Flanders, this concerns about 33500 ha of Scots pine (*Pinus sylvestris*) and 11500 ha of Corsican pine (*Pinus nigra var. laricio*) stands on sandy soils (Geudens et al. 2006) of a total Flemish forest area of 146000 ha. This thesis focuses on the ecoregion of the Kempen (see Fig.1.11), covering the Northeast of Flanders and containing 90% of the Flemish coniferous plantations with 35% of its area being Scots pine stands and around 10% Corsican pine stands. The distribution of pine plantation in Flanders is obviously linked to poor sandy soils (Geudens et al. 2006).

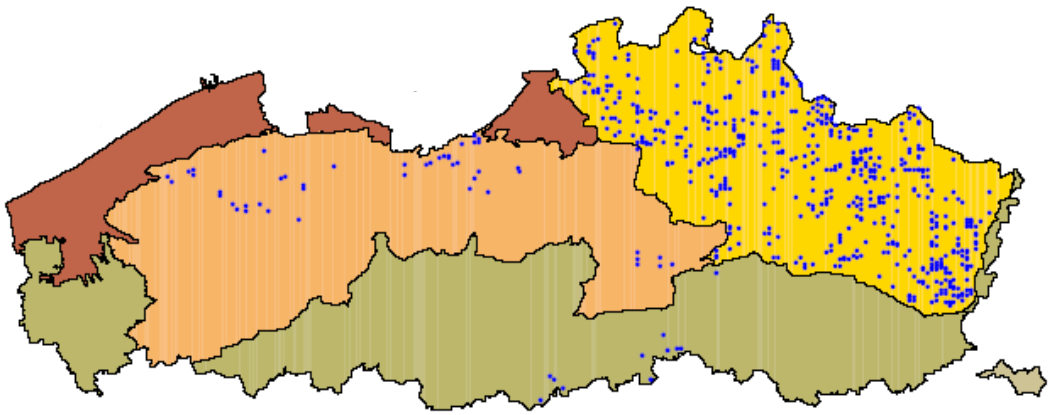

Fig.1.11: Distribution of Scots pine stands in Flanders. The Kempen ecoregion (in yellow), covers the northeast part of Flanders (Adopted from Geudens et al. 2006)

Fig. 1.12 gives a schematic overview of the structure of the thesis and the questions it aims to answer. The first part of the thesis handles on different aspects of acidification and eutrophication of Flemish forest ecosystems. In Chapter 2 we aim to find out if sandy forested podzol soils are acidified during the past 50 years. We furthermore discuss in this chapter whether the average annual acidification rate accelerated or slowed down between 1985 and 2000 compared to the period 1950-1985, considering the significant emission reductions

during the period 1990-2000. In Chapter 3 we study the relationship between atmospheric throughfall deposition load and soil acidification. In this chapter we also study the spatial variability of atmospheric throughfall deposition input, by comparing inputs at the forest edge to the forest interior. In Chapter 4 we furthermore estimate the relevance of considering this higher deposition load in forest edges for calculating exceedance of critical loads for nitrogen and potentially acidifying deposition.

In part II of the thesis, we study the effects of both the forest type and the deposition load on ion throughfall deposition and seepage to groundwater. Chapter 5 compares several vegetation types (plantations of *Pinus sylvestris*, plantations of *Pinus nigra*, mixed deciduous forest with *Betula pendula* and *Quercus robur* and pure and grassed heathland ecosystems) in an area with relatively low deposition load. In Chapter 6, a stand of *Pinus nigra* is compared to a stand of *Betula pendula* in an area with high emissions of ammonia. In Chapter 7 we furthermore aim to find out whether the found results in chapters 5 and 6 are to generalize to other forest types and deposition regimes. The answers of the formulated questions are given in the summarizing conclusions in Chapter 8. In this chapter, we also discuss the risks and consequences of acidification and eutrophication of forest ecosystems, and examine the possibilities of forest conversion for forest ecosystem recovery.

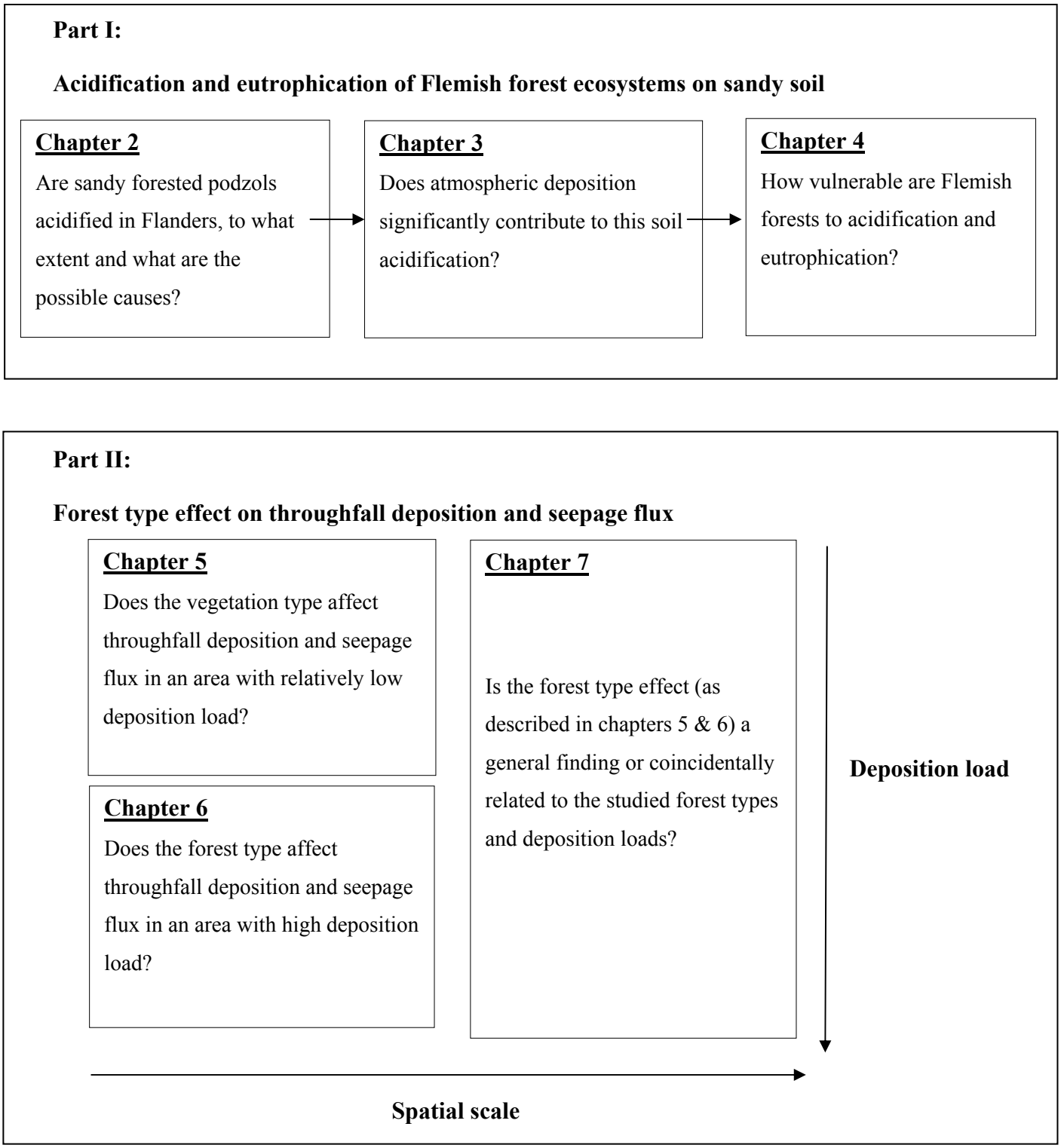

Fig. 1.12: Schematic overview of the thesis

2 Acidification of forested podzols in Flanders (northern Belgium) during the period 1950-2000

After: De Schrijver A., Mertens J., Geudens G., Staelens J., Campforts E., Luyssaert S., De Temmerman L., De Keersmaeker L., De Neve S. & Verheyen K. (2006). Acidification of forested podzols in northern Belgium during the period 1950-2000. Science of the Total Environment 361: 189-195.

2.1 Abstract

Acidification of forest soils in Europe and North America has been an important concern over the last decades. The last area-covering survey of forest soil acidification in Flanders (northern Belgium) goes back to 1985 (Ronse et al. 1988), and highlighted a significant acidification of the upper layer (0.3-0.4 m) of forested podzols during the period 1950-1985. The present study aimed to assess (1) to what extent further acidification of forested podzols occurred during the period 1985-2000 at different depths and (2) whether the average annual acidification rate accelerated or slowed down between 1985 and 2000 compared to the period 1950-1985.

Average soil pH-KCl values of podzols in northern Belgium dropped during the period 1985-2000. This decline extends to a depth of about 50 cm but was most pronounced and significant in the A horizon. In the A_0, A_1 and A_2 horizons, average pH dropped with 0.2, 0.3 and 0.1 units and in the B_{ir} and C horizons with 0.1 units. No change in average pH value occurred in the B_h horizon. Average annual acidification rate of the A_1 horizon was significantly higher in the period 1985-2000 than in the period 1950-1985.

Changes in pH occurred in the entire soil profile during the period 1950/67-1985 likely because sulphate was the major form of potentially acidifying deposition before 1985. After 1985, sulphur deposition decreased with more than 50% in northern Belgium. In contrast, ammonium deposition almost doubled between 1950 and 1980, which may explain why soil acidification between 1985 and 2000 has been restricted to the upper soil horizons since acidification as a consequence of ammonium uptake by vegetation and nitrification reactions is mainly restricted to the upper soil horizons.

2.2 Introduction

Acidification of forest soils in Europe and North America has been an important concern over the last decades. A large number of recent studies report a significant forest soil acidification over the past 50 years (e.g. Ahokas 1997, Drohan ad Sharpe 1997, Dai et al. 1998, Hovmand and Hansen al. 1999, Van der Salm and De Vries 2000, Miller et al. 2001, Heisner et al. 2003). Podzol soils, which are naturally acid and depleted in nutrients, are particularly sensitive to acidification because of the low cation-exchange capacity and the relatively small amounts of weatherable minerals (Wiklander and Anderson 1972). Such sandy, podzol soils are widespread in Flanders, the region of the present study. In addition, acidification of these podzols may be stimulated by the prevailing cover of pine plantations, past forest management practices and intensive cattle breeding in the studied region.

First of all, the prevailing forest types on podzol soils in the study area are plantations of Scots pine (*Pinus sylvestris* L.) and Corsican pine (*Pinus nigra ssp. laricio* Maire) (Afdeling Bos en Groen, 2001). Plantations of coniferous species are known to lead to notable modifications of the soil (Ovington 1955, Challinor 1968, Perala and Alban 1982, Nihlgard 1972, Binkley and Valentine 1991, Berger et al. 2004). Compared to broadleaved species, coniferous litter is more resistant to biological degradation (Howard and Howard, 1990, Johansson, 1995) and has lower amounts of exchangeable base cations (Zinke 1962, Muys et al. 1992, Finzi et al. 1998, Reich et al. 2005). Furthermore, conifers are found to be more efficient in collecting particles and cloud droplets from the atmosphere, enhancing potentially acidifying deposition (Brown and Iles 1991, Draaijers 1993, De Schrijver et al. 2004, De Schrijver et al in press).

A second factor that can affect soil acidification under the Flemish pine plantations is the past management, including clear cutting at 30-50 year intervals, total tree use and litter raking (Burny 1999). All mentioned management practices are known to increase soil acidity especially because of base cation removal (Nilsson et al. 1982, Van Breemen 1988, Johnson et al. 1991, Dahlgren and Driscoll 1994, Prietzel et al. 1997, Adams et al. 2000).

Thirdly, in the middle of the 20th century, intensive livestock husbandry has developed as the main agricultural activity on sandy soils in Flanders, causing high emissions of ammonia. Average potentially acidifying deposition in Flanders is among the highest in Europe (UNECE-EC 2003) and in spite of an important decrease in potentially acidifying deposition (-33% in 2004 in comparison with 1990), all medium and long-term targets formulated in the

Protocol of Göteborg and the European NEC directive are still exceeded (Van Avermaet et al. 2006).

The last area-covering survey of forest soil acidification in Flanders goes back to 1985 (Ronse et al. 1988). This survey highlighted a significant acidification of the upper layer (0.3-0.4 m) of forested podzols during the period 1950-1985. In 1985, most values of the mineral soil were situated in cation exchange buffer range. We hypothesized that, as a consequence of the high deposition load, further soil acidification did take place.

The first aim of the present study was to assess to what extent further acidification of forested podzols occurred during the period 1985-2000 at different depths. Our second aim was to find out whether the average annual acidification rate accelerated or slowed down between 1985 and 2000 compared to the period 1950-1985, considering the significant emission reductions during the period 1990-2000.

2.3 Material and methods

2.3.1 Data collection

To investigate soil acidification, about 100 not limed soils were sampled in 1985 to determine soil pH (Ronse et al. 1988). These values were compared to pH values measured in the period 1950-1967 within the framework of the Belgian soil cartography (Ronse et al. 1988). On both sampling occasions, spatial coordinates, topography, vegetation, and soil profile characteristics (colour, thickness and texture of the horizons) were reported. Soil was sampled from the different horizons in a profile pit.

In 2000, the subset of forested podzols (originally 67 of 100 locations) from Ronse et al. (1988) was reconsidered. Only 33 of the former 67 forested sites on sandy podzols were encountered in an undisturbed state. 34 sites were deforested, built-over or cultivated. Four of the remaining forested sites were cut and replanted through 1985 to 2000. These sites were not considered in the analysis since harvesting operations lead to more favourable soil moisture and temperature conditions for growth of micro-organisms, decomposition of resistant litter can be accelerated, and immobilized cations can be released, influencing soil pH (Hendrickson et al. 1989, Bock and Van Rees 2002, Pennock and Van Kessel 1997). Of the 29 remaining plots, 22 were situated under coniferous forest and 7 under deciduous forest.

Based on the available information, the sampling sites were relocated. However, digging the soil profile pits in 1988 disturbed the original sample points. Therefore, we took five soil samples in undisturbed soil with a gouge auger. One sample was taken as close as possible to the original site and four additional samples were taken at a 2 m radius around it, one in each cardinal direction. To assess local variability of soil pH, samples were not pooled but stored and analysed separately.

Each auger sample was divided into soil horizons, and subsamples of each horizon were taken after measuring the thickness of each horizon (Table 2.1). Within the eluvial or A horizons, distinction was made between the upper mineral horizon A_1 and the leached mineral horizon A_2. Iluvial or B-horizons were those enriched with humus (B_h) or iron (B_{ir}). The parent material was indicated as the C horizon.

Table 2.1: Mean depth (cm) of the soil horizons of podzols (n = 29)

Level	Horizon	Depth
0	A_0	-
1	A_1	0-11
2	A_2	11-24
3	B_h	24-35
4	B_{ir}	35-44
5	C	> 44

2.3.2 Sample preparation and chemical analyses

Samples were dried for 48 h at 50 °C before sieving (soil samples) or grinding (litter samples). Soil pH-KCl was measured since this potential acidity is less dependent on the soil moisture content and consequently less variable than pH-H_2O (Thomas 1996). The pH was measured using a glass electrode (Orion, model 920A) after extracting 8 g air-dried soil or litter with 20 ml KCl solution (1 M) during 15 minutes, similarly to the previous research of Ronse et al. (1988).

2.3.3 Quality control

The quality of chemical analyses was checked by including repeated measurements of reference samples. Each 15 samples, a certified sample of pH=6 was analyzed. The results

were rejected if the pH value was higher than 6.02 or lower than 5.98. Then, the previous 15 samples were re-analyzed after calibration.

2.3.4 Statistical analysis

For further statistical analyses, the 5 replicate pH values were averaged for each soil horizon of each sampling point since standard deviation was very low (coefficient of variation = 4.23%). Before averaging, pH values were converted into H^+ concentrations. Average soil pH of each soil horizon was then obtained by calculating the –log(average H^+-concentration).

To compare the average soil or litter H^+ concentrations in 1950/67, 1985 and 2000, Wilcoxon signed rank tests were performed (Neter et al. 1996). We choose this non-parametric test because the differences in H^+-concentrations between years for each horizon did not meet the conditions of normality. For the set of three pairwise comparisons, a Bonferroni correction was applied if differences in H^+ concentrations were compared between years. In that case, a confidence coefficient of 0.017 was obtained through division of the family level 0.05-confidence coefficient by three (Neter et al. 1996).

To compare the average annual acidification rate between the periods 1950/1967-1985 and 1985-2000, the data were expressed in terms of increase of H^+-concentration per gram of soil and per year. Therefore, for each sampling point, the H^+-concentrations per gram of soil were divided by the exact time span (number of years) between measuring dates. For this dataset, the conditions of normality were met and the average annual acidification rate between both periods was compared using a paired samples t-test (Neter et al. 1996). Historical data of litter pH-KCl were only available for the year 1985, so no comparison of the average annual acidification rate between periods was possible for the litter layer.

All statistical analyses were made with SPSS 11.0 for Windows.

2.4 Results

2.4.1 Comparison of pH values and H^+-concentrations between sampling years

The average pH-KCl of the humus layer (A_0) decreased from 3.0 to 2.8 (Fig. 2.1) between 1985 and 2000. The pH of the A_1 horizon declined 0.1 unit between 1950/67 and 1985, and 0.3 units between 1985 and 2000. In contrast to the A_1 horizon, the pH in the A_2 horizon declined more between 1950/67 and 1985 (0.3 units) than between 1985 and 2000 (0.1 unit).

Similarly to the A_2 horizon, the pH of the B horizons and the C horizon declined more between 1950/67 and 1985 (Fig. 2.1).

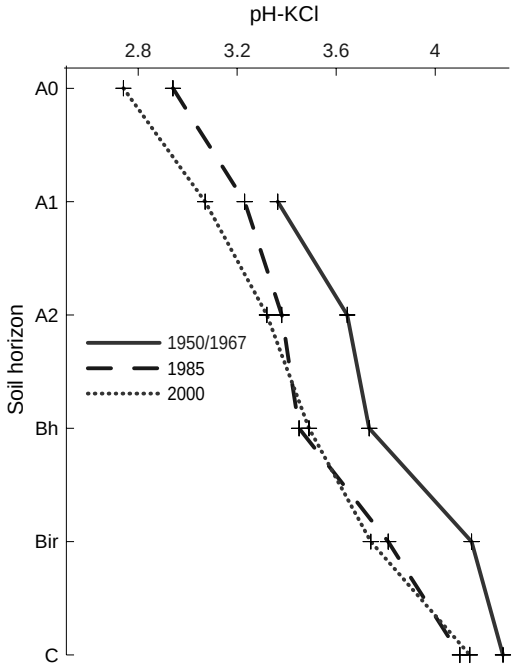

Fig. 2.1: Evolution in time of pH-KCl in different horizons of 29 podzol soils

The H^+-concentrations in 2000 significantly increased compared to 1950/67 throughout the complete mineral soil profile (Table 2.2 and Table 2.3). Between 1985 and 2000, H^+ concentrations increased significantly in the three A-horizons (Table 2.3). In the deeper horizons, no significant change in H^+ concentration was detected in this period. Changes in H^+ concentrations did not differ between coniferous and deciduous plots (numbers not shown).

Table 2.2: Average H^+-concentrations and their standard deviation (10^{-6} mol.g^{-1} soil) in different horizons of 29 podzol soils

Level	1950/67	1985	2000
A_0	-	2.56 ± 1.97	4.25 ± 2.07
A_1	1.06 ± 1.12	1.32 ± 0.83	2.27 ± 1.06
A_2	0.48 ± 0.53	0.95 ± 0.86	1.21 ± 1.02
B_h	0.45 ± 0.48	0.88 ±1.16	0.82 ± 0.77
B_{ir}	0.16 ± 0.06	0.36 ±0.86	0.48 ± 0.67
C	0.14 ± 0.17	0.21 ±0.19	0.18 ± 0.14

Table 2.3: Significance level (p) of the difference in H^+ concentrations between the sampling years 1950/67, 1985, and 2000, as given by a Wilcoxon signed rank test. Average soil horizon H^+ concentrations were significantly different at the 95% confidence level (indicated by *) when $p < 0.017$, because of the Bonferroni correction for multiple comparisons.

Level	1950/67-1985	1985-2000	1950/67-2000
A_0	-	0.005*	-
A_1	<0.001*	<0.001*	<0.001*
A_2	0.003*	0.008*	<0.001*
B_h	0.006*	0.581	0.003*
B_{ir}	0.046	0.109	0.002*
C	0.021	0.023	0.003*

2.4.2 Comparison of average annual acidification rate during the periods 1950/67-1985 and 1985-2000

The average annual acidification rate in the A_1-horizon increased from $8 \pm 48 \times 10^{-9}$ mol.g^{-1}.yr^{-1} between 1950/67-1985 to $63 \pm 56 \times 10^{-9}$ mol.g^{-1}.yr^{-1} between 1985-2000 (Fig. 2.2). According to a paired t-test, which takes the difference in acidification rate of individual sample points into account, this increase was highly significant. This means that the acidification process in the A_1-horizon significantly accelerated during the period 1985-2000 compared to 1950/67-1985.

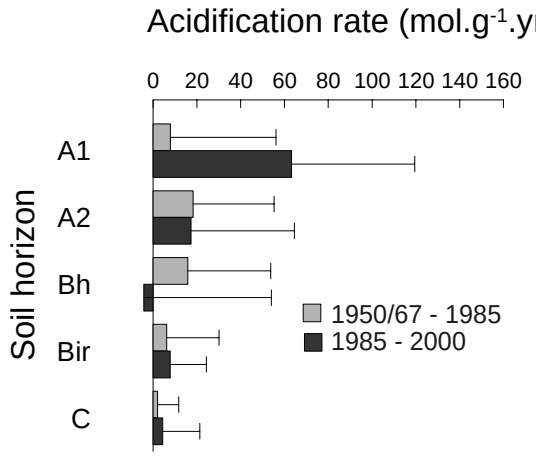

Fig. 2.2: Average annual acidification rate (mol.g^{-1}.yr^{-1}) with standard deviation bars in positive direction during the periods 1950/67-1985 and 1985-2000

In the A_2-horizon, the acidification rate remained unchanged between 1950/67 and 2000 (18 ± 37 and 17 ± 47 x 10^{-9} mol.g^{-1}.yr^{-1} for both periods, respectively). In the B_h, B_{ir} and C horizon, the differences in the acidification rate were also not significant.

2.5 Discussion

2.5.1 Which buffer systems were/are responsible for acid neutralisation?

In the present study, samples of 29 forest soils were analysed on pH-KCl giving insight in the potential acidity of the soil. No analyses on actual acidity (pH-H_2O) were performed since the high dependence on soil moisture content can affect interpretation of temporal changes (Thomas 1996). Based on pH-H_2O values, Ulrich (1991) distinguished different buffer ranges for the interpretation of pH values. PH-H_2O and pH-KCl values of a soil sample on average differ about 1 unit (Thomas 1996), but the actual difference depends on its cation exchange capacity, aluminium saturation and carbon content. To be able to interpret our results in terms of buffer ranges (that are expressed in pH-H_2O units, see Table 1.5), we used an average difference of 0.7 units, based on the observed average difference between pH-H_2O and pH-KCl along the transect in chapter 3 (see §3.4). From this rather rough estimation, it can be concluded that in 2000 the A_1 horizons in most surveyed plots were situated in the aluminium or in the aluminium/iron-buffer range. This means that in 2000 some of the studied podzol soils have moved into the range where besides Al^{3+}, also Fe^{2+} comes into solution and can leach to the groundwater. The average pH of the A_2 horizon is since 1985 situated in the aluminium buffer range, while in 1950 these horizons and the B horizons during the total measuring period were on average still within the cation exchange buffer range (which ranges from pH-H_2O 4.2 to 5). At some locations, the C-horizons were probably still in the silicate buffer range. Evidence for the shift of forested topsoils to the aluminium buffer range can be found in the high concentrations of aluminium in the soil solution found by Neirynck et al. (2002) and De Schrijver et al. (2004).

2.5.2 What were/are possible causes for soil acidification?

Acidification of forest soils is a result of natural soil forming processes, forest growth and anthropogenic activities as input of potentially acidifying deposition. The average internal proton production for six podzolic forest soils in the Netherlands, mainly as a consequence of organic acid deprotonation, was calculated to range from 0.1 to 0.9 kmol.ha^{-1}.yr^{-1} (De Vries

and Breeuwsma 1985). Proton production as a consequence of forest growth depends on the type of harvesting and was reported to vary from 0.5 to 2.0 kmol.ha^{-1}.yr^{-1} (Ulrich and Matzner 1983, Van Breemen et al. 1983) in case of total tree use and 0.5 to 0.6 kmol.ha-1.yr^{-1} (Ulrich 1983c) in case of conventional harvesting, in which only stems are removed. In northern Belgium, potentially acidifying deposition in 2000 was found to be on average 4.4 kmol.ha^{-1} (Van Avermaet et al. 2006). Summarizing it can be assumed that soil acidification resulting from atmospheric deposition clearly exceeds acidification by internally generated protons on this type of soils. The importance of potentially acidifying deposition for the acidification of forest soils was also confirmed by Bredemeier (1989), who calculated the internal generation of acidity to account for less than 30% of total proton load.

2.5.3 Why is the acidification profile different between two measuring periods?

Changes in pH occurred in the entire soil profile during the period 1950/67-1985, while the main changes between 1985 and 2000 were restricted to the A horizons. We hypothesize that this observation can be related to the different composition of the potentially acidifying atmospheric deposition during the last 50 years. In the eighties, sulphur (S) emissions were generally considered the most important cause of potentially acidifying deposition. However, since then S emissions and thereby S deposition have constantly decreased over large parts of Europe, whereas NH_3 emissions mostly stayed constant or even increased (de Vries et al. 2003). This general European pattern largely holds for the region of the present study. Emissions of SO_2 in Belgium in 1970 were twice as high as in 1950 and in 1985 (Mylona 1996). Furthermore, S deposition decreased with about 50% in northern Belgium between 1990 and 2000 (Van Avermaet et al. 2006). Deposition of ammonium in Belgium almost doubled between 1950 and 1980 (Asman et al. 1988) and decreased (-8%) between 1990 and 2000 (Van Avermaet et al. 2006). In 2000, the average potentially acidifying deposition in Flanders amounted to 4396 mol.ha^{-1}.yr^{-1}, of which 1173 mol.ha^{-1}.yr^{-1} came from SO_x, 1258 mol.ha^{-1}.yr^{-1} from NO_y, and 1965 mol.ha^{-1}.yr^{-1} from NH_x (NH_4^+ deposition was converted to deposition of H^+ assuming complete nitrification in the soil) (Van Avermaet et al. 2006).

The acidifying effect of the potentially acidifying pollutants NO_3^- and SO_4^{2-} is largely dependent on their chemical composition when deposited. When deposited as a salt (e.g. KNO_3 or $CaSO_4$), no net proton production will occur in the soil (Binkley and Richter 1987), while deposited as an acid (HNO_3 and H_2SO_4), acidification occurs when nitrate or sulphate are seeped from the system with an accompanying base cation. Both HNO_3 and H_2SO_4 are

strong acids with low affinity for H^+. When entering a soil, the protons are released very quickly and seepage of nitrate occurs primarily with multivalent cations as Mg^{2+}, Ca^{2+} and Al^{3+}. Nitrate is not strongly adsorbed to the soil complex and is therefore very mobile, while sulphate can be adsorbed or desorbed from the soil, consuming or generating protons (see also §1.2.2). The fact that S deposition was the dominant potentially acidifying pollutant can explain why the average pH dropped in the complete soil profile during the period 1950/67-1985, since neutralisation of the seeping sulphate occurs throughout the whole soil profile (Ulrich 1983b).

During the period 1985-2000, with NH_4^+ as dominant potentially acidifying pollutant, soil acidification mainly occurred in the upper soil horizons. The acidifying effect of ammonium is more complex. When deposited as NH_4^+, it can be taken up by plant roots ($+1H^+$), assimilated into organic matter ($+1H^+$), fixed at the cation exchange complex (CEC) ($1H^+$) or oxidized to nitrate (nitrification, $+2H^+$). All these reactions are generating protons and mainly occur in the upper soil horizons, since NH_4^+ is not mobile at all. This is confirmed by the low amounts of NH_4^+ detected in the soil solution at soil horizons deeper than 10 cm.

2.5.4 How much buffer capacity is still left?

Mineral weathering can help offset soil acidification - either directly in the upper soil horizons or indirectly due to weathering in lower soil horizons and redistribution due to plant uptake and litterfall. Our results can be related to the study of Van Ranst et al. (2002), who thoroughly studied the current acid neutralizing capacity (ANC) represented by the cations potassium, calcium, magnesium, sodium and aluminium of the litter layer and the upper 10 cm of the mineral soil of two podzols under coniferous forest in northern Belgium. The soil, the vegetation and the deposition load of both plots are representative for the conditions of most of the plots in our survey. When the total ANC values of the forest floor (67 and 96 $kmol.ha^{-1}$ for both plots, respectively) and the upper 10 cm of the mineral soil (1147 and 871 $kmol.ha^{-1}$) (Van Ranst et al. 2002) are compared with the average potentially acidifying loadings in both plots (between 4.4 and 4.7 $kmol.ha^{-1}.yr^{-1}$), and supposing that all acid input is buffered in the mentioned upper soil horizons, only 205 to 276 years of buffering capacity of the upper 10 cm is still left. In reality, this period will be longer than calculated since part of the acid will also be buffered in deeper horizons, for which no ANC-calculations were performed.

The critical load for protecting the acid buffering potential of the soil was exceeded in the majority of Flemish forest ecosystems in 2000 (Van Avermaet et al. 2006). This clearly indicates the dramatic condition of northern Belgian forest ecosystems and points out the urgent need for further drastic reduction emissions in the near future.

2.6 Conclusions

Between 1950 and 2000, soils acidified significantly in the upper 35 cm of the mineral soil profile. In 1985 mineral soils were still in the cation exchange buffer range. In 2000 soils did further acidify significantly in the upper 25 cm of the mineral soil and the upper 10 cm of the mineral soil profile evolved towards the aluminium buffer range. In the upper mineral soil horizon (A_1), the annual acidification rate differed significantly between the two measuring periods, while in the deeper horizons no differences were found. Changes in pH occurred in the entire soil profile during the period 1950/67-1985, while the main changes between 1985 and 2000 were restricted to the A horizons. We hypothesize that this observation can be related to the different composition of the potentially acidifying atmospheric deposition during the last 50 years, which evolved from being dominated by SO_4^{2-} to NH_4^+.

3 Soil acidification along an ammonium deposition gradient in a Corsican pine stand in northern Belgium

After: De Schrijver A., Nachtergale L., Roskams P., De Keersmaeker L., Mussche S. & Lust N. (1998). Soil acidification along an ammonium deposition gradient in a Corsican Pine stand in northern Belgium. Environmental Pollution 102: 427-431.

3.1 Abstract

In a homogeneous Corsican pine (*Pinus nigra ssp laricio* Maire) forest situated in a region of intensive live stock breeding, the effect of different NH_x doses on soil acidification was studied. Throughfall collectors were placed along a transect of 1.4 km. Along this transect, three zones of significantly differing NH_4^+ throughfall depositions were identified: a forest edge zone of 270 m at the south-western border, the forest interior, and a forest edge zone of 150 m at the north-eastern side of the forest.

The differences in throughfall deposition of NH_4^+, and probably also of nitrate and sulphate, were clearly reflected in the soil pH of the topsoil (0-10 cm). The Pearson correlation coefficient between the ln-transformed NH_4^+ throughfall deposition and the pH-H_2O amounted to $r = -0.572$ ($p < 0.001$), which indicates the negative significant relationship between both parameters. The topsoil was very acid along the complete length of the transect: pH-H_2O, which is a measure for actual acidity, varied from 3.05 (edge) to 3.80 (centre), while pH-KCl values varied from 2.35 (edge) to 2.80 (centre). In the forest interior, soils are in the aluminium buffer range, while at the forest borders soils already moved on to the iron buffer range.

3.2 Introduction

Over 60% of the Flemish (northern part of Belgium) forest area is located on poor sandy soils which are mainly afforested with Scots pine (*Pinus sylvestris*) and Corsican pine (*Pinus nigra ssp. laricio* Maire). Because of the low fertility of these soils characterised by low buffering capacity and originally low levels of available nitrogen, the main agricultural practice in these regions is semi-industrial live-stock breeding. This live-stock production is the dominant source of atmospheric NH_x and the considerable rise of it during the last decades has caused a significant increase in NH_x depositions (Dams et al. 1996).

The deposition of NH_x on pine forests is far higher than the national average. This is not only due to the proximity of important sources of NH_x, but also because of the higher filtering capacity of coniferous tree canopies in comparison to other vegetation types (Brown and Iles 1991). Furthermore, forest edges disturb the vertical wind profile and so cause higher air turbulence and inflow (Draaijers et al. 1988, 1994). Dry deposition velocities near forest edges are substantially increased and consequently trees in forest edges catch much more air pollutants than trees in the middle of the forest. The enhancement of dry depositions in forest edges strongly depends on wind velocity and wind direction.

Ammonium deposited on forest soils has different pathways: consumption by vegetation, adsorption on the cation exchange complex of the forest floor and mineral soil, assimilation into organic matter and nitrification by bacterial activity (van Breemen et al. 1988), all resulting in proton release, and so causing acidification of the upper soil horizons. Subsequently high amounts of NO_3^- can seep beneath the rooting zone and pollute the groundwater. Processes like soil acidification and nitrate seepage can be more intense in forest edges (Draaijers et al. 1988).

In this chapter, we hypothesize that higher amounts of ammonium throughfall deposition are reflected in a lower soil pH. Therefore, an experiment was started in a homogenous Corsican pine forest on sandy soil situated in a region of intensive livestock. The methodology was based on three principles: (1) ammonium throughfall deposition is high due to the vicinity of intensive livestock, (2) pine forests are effective filters for air pollutants and (3) at forest edges strong air turbulences develop, so ammonium throughfall deposition is higher than in the centre of the forest.

3.3 Materials and methods

3.3.1 Study site

The experiment was set up in the state forest of Ravels in the northern part of Belgium (51°22'N, 5°2'OL). The forest covers about 800 hectares and is mainly composed of pine stands (*Pinus sylvestris* and *Pinus nigra ssp. laricio* Maire) established on former heathland in the beginning of the 20th century. The forest is partly located on the High Campine plateau and ranges in elevation from 30 to 35 meters. The Campine plateau originates from a mixture of tertiary sands and gravel-rich sands deposited by the river Meuse. During the Pleistocene these sands were covered by aeolian sand deposits. The coarse sandy soil has a massive, compacted and cemented spodic B horizon and is classified as a Haplic Podzol (FAO 1988). The state forest is surrounded by several sties with intensive livestock breeding (pigs, cows and chickens).

3.3.2 Data collection

A transect of ±1.4 km was established from the south-western to the north-eastern part of the forest (Fig. 3.1). Along this transect, soil characteristics (texture, profile development and drainage class) and stand characteristics (*Pinus nigra ssp. laricio* Maire; 70 years) were quite similar. Every 25 meter along the transect throughfall water was collected monthly between May and November 1996. On each point four two-litre recipients fitted with funnels (∅ 14 cm) were placed below ground to avoid growth of algae. A nylon wire mesh was placed in the funnel to prevent contamination by large particles. The throughfall water was collected solely under tree canopies and not in open spots. Stemflow water was not collected because of its low contribution to nutrient fluxes in pine stands (less than 0.6% of bulk precipitation; Neirynck 1996). To obtain a sample representative for each distance, the collected water of the four recipients was mixed. The samples were analysed on NH_4^+ with a NH_4^+ specific electrode (ORION). Because of the prevailing westerly and south-westerly wind directions in Flanders, high NH_x depositions were expected near the south-western border of the forest.

Every 25 meter, eight soil samples of the upper mineral soil (0-10 cm) were taken, sieved, mixed and analysed on pH-H_2O and pH-KCl with a specific electrode (ORION).

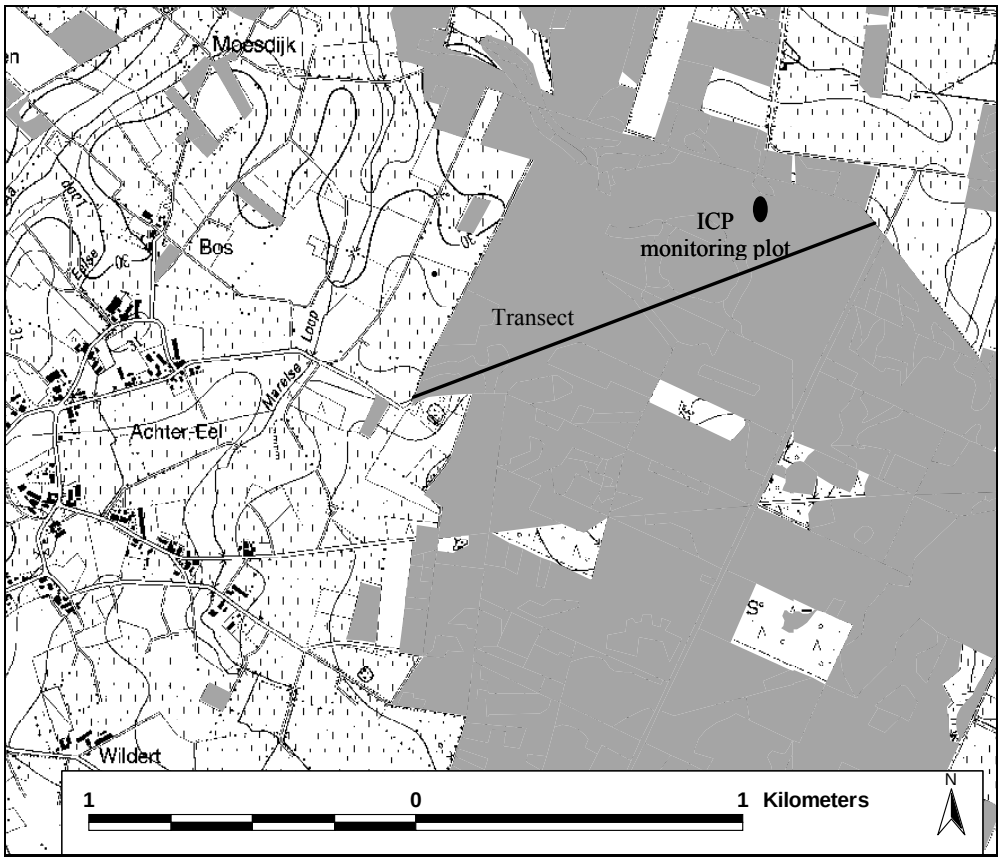

Fig. 3.1: Map of the northern part of the state forest of Ravels with indication of the study transect and the ICP forest monitoring plot

In the vicinity of the transect, at about 1000m from the SW-border, a permanent ICP Forest monitoring plot (International Cooperative Programme on assessment and monitoring of air pollution effects on forests) of 25 acres is located (Fig. 3.1). Within this plot of Corsican pine, quality and quantity of different water fractions is permanently (fortnightly) monitored. Among other water fractions, throughfall water is sampled using 10 randomly distributed bulk collectors. The funnels (∅ 15 cm) of these collectors are placed at 1 meter height in order to avoid the influence of herbs and the polyethylene recipients are placed below ground level. Also bulk deposition is collected using 4 bulk collectors in a meadow outside the forest. The collectors are sampled and replaced by distilled water rinsed collectors fortnightly. Analyses are conducted on pH, K^+, Ca^{2+}, Mg^{2+}, Na^+, NH_4^+, NO_3^-, SO_4^{2-} and Cl^-.

3.3.3 Data analysis

In order to distinguish zones with differing NH_x deposition along the transect, the method designed by Beier and Gundersen (1989) was used. The fluxes were ln-transformed and plotted against distance to the forest edge. Subsequently zones were separated showing a linear relationship between distance and the ln-transformed fluxes. The zones under influence of higher turbulences, the forest edge zones, show a linear relationship with a slope < 0. The zone of the forest interior can be characterized by an absence of relationship between distance from the forest edge and the ln-transformed NH_4^+ deposition. The depth of the forest edge zones can then be designated as the X-axis (distance) value of the intersection of both regression lines (Wuyts et al. submitted).

After testing the preconditions of normality and homoscedasticity, pH values and ln-transformed NH_4^+ throughfall deposition values were compared between the three distinguished zones - an edge zone at the SW border, a NE edge zone and the so-called 'forest interior zone' - by means of a one-way Anova test. The relation between soil acidification and NH_4^+ throughfall deposition was studied by calculating the Spearman correlation coefficient between the ln-transformed NH_4^+ throughfall flux and the pH-H_2O values.

3.4 Results

Ammonium was the dominant inorganic nitrogen compound in both bulk and throughfall deposition (Table 3.1) measured in the permanent ICP monitoring plot. The contributions of ammonium, sulphate and nitrate to the potentially acidifying throughfall deposition amounted to respectively 50%, 35% and 15 % on a total of 3.27 keq.ha^{-1}.yr^{-1}. Strong and significant correlations were found between ammonium and respectively nitrate and sulphate concentrations in throughfall water (Fig. 3.2).

Table 3.1: Annual values of bulk and throughfall deposition in the permanent monitoring plot in Ravels in 1996 in kmol.ha^{-1}.yr^{-1} except for H^+ in mol.ha^{-1}.yr^{-1}

	NH_4^+-N	**NO_3^--N**	**SO_4^{2-}-S**	**Cl^-**	**K^+**	**Ca^{2+}**	**Mg^{2+}**	**Na^+**	**H^+**
Bulk deposition	1.40	0.21	0.20	0.56	0.06	0.12	0.03	0.50	0.21
Throughfall deposition	2.02	0.49	0.38	0.73	0.37	0.10	0.04	0.63	0.49

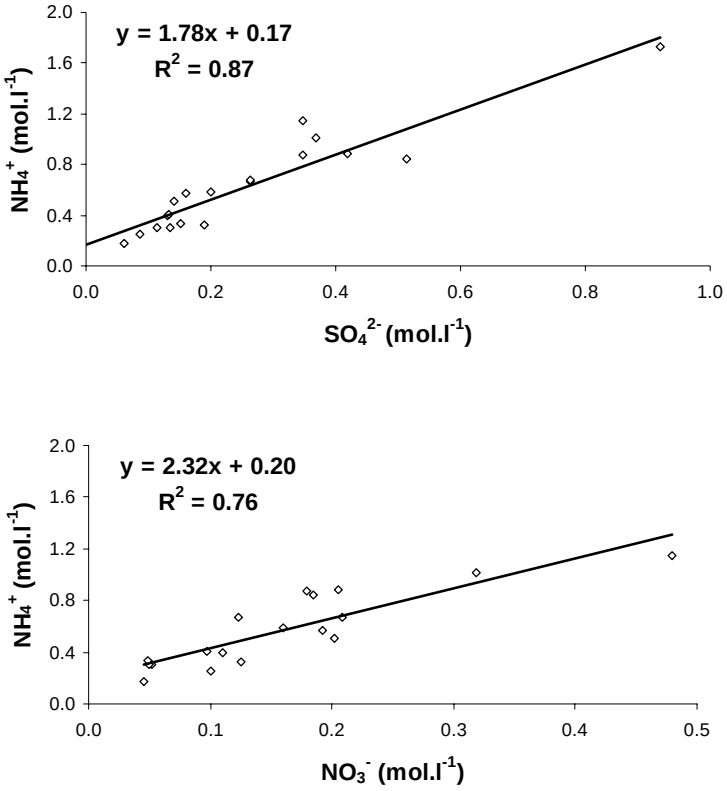

Fig. 3.2: Correlation between NH_4^+ versus respectively SO_4^{2-} and NO_3^- concentrations ($mol.l^{-1}$) in throughfall water

In the south-western forest edge zone of 270 m (Fig. 3.3), NH_4^+ throughfall deposition was found to be significantly ($p<0.001$) higher than in the centre of the forest. The outer point of the south-western (SW) edge of the forest receives 2.4 times more NH_4^+ throughfall deposition compared to its average value in the forest interior (Fig. 3.4). For the north-eastern (NE) forest edge, we could not distinguish a realistic forest edge zone by means of the method of Beier and Gundersen (1989) (see §3.3.3) because of the weak decrease in NH_4^+ throughfall deposition. We visually determined a 100 m edge zone at the NE side (Fig. 3.3), with smaller (about 1.3 times) enhancements of NH_4^+ throughfall deposition at the outer point of the edge (Fig. 3.4).

NH_4^+ throughfall deposition values were significantly different in the three distinguished zones: the SW forest edge zone of 270m, the NE forest edge zone of 150m and the forest interior. Climatic data showed south-western winds to be dominant during the sampling period (May - November 1996).

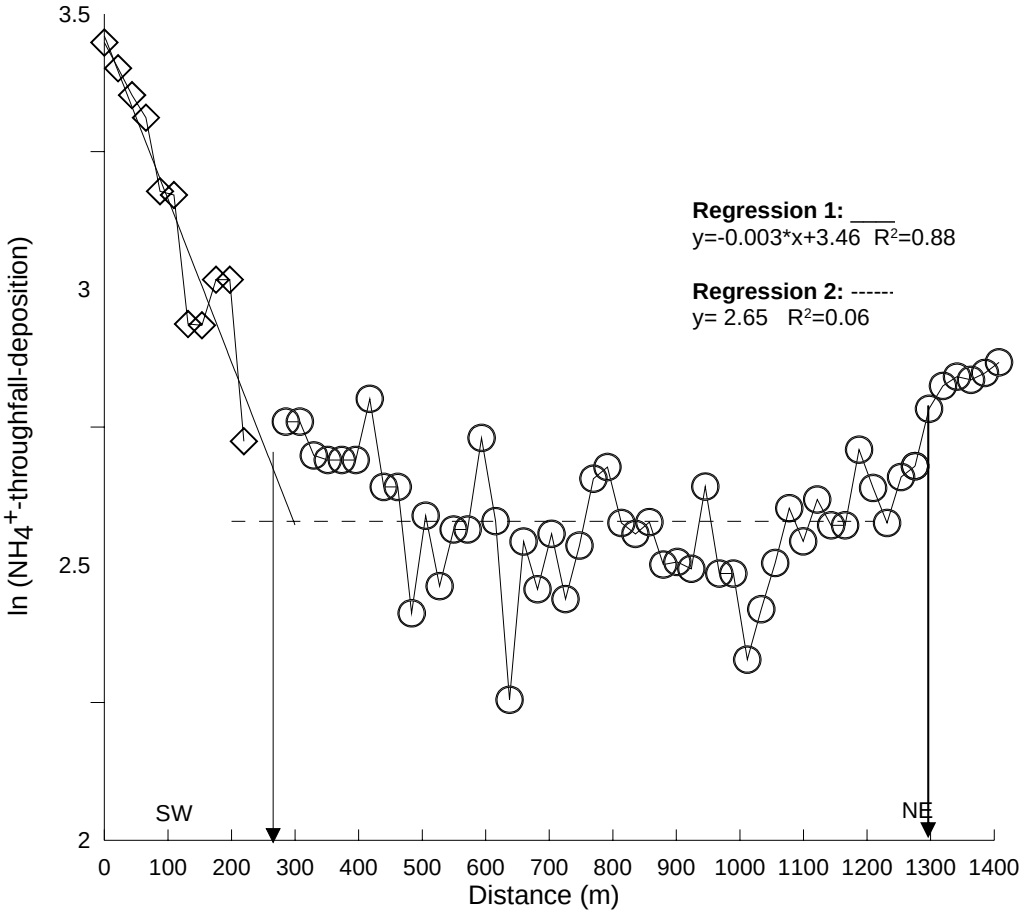

Fig. 3.3: Ln-transformed NH_4^+ throughfall values along the 1400m long transect. Regression lines calculated according to the method of Beier and Gundersen (1989).

Extremely low soil-pH values were measured in both forest edges, especially in the south-western edge where pH-H_2O-values vary from 3.05 to 3.15 (Fig. 3.4) and pH-KCl values range between 2.35 and 2.55. In the centre of the forest, pH-H_2O and pH-KCl reach values of respectively 3.8 and 2.8. We compared the average pH values between the three distinguished forest zones and found significantly ($p<0.01$) different pH values in the two forest edge zones compared to the forest interior.

The Spearman correlation coefficient between the ln-transformed NH_4^+ throughfall deposition and the pH-H_2O values amounted to $r = -0.668$ ($p < 0.001$), which indicates the negative and significant relationship between both parameters (Fig. 3.5).

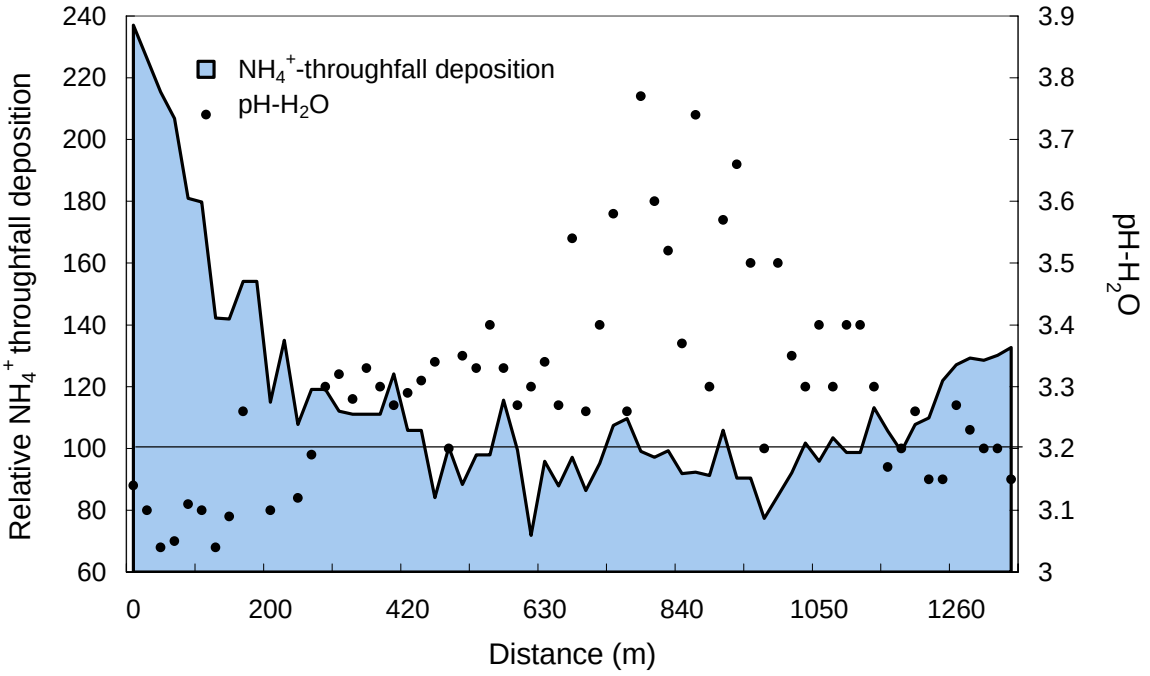

Fig. 3.4: Throughfall deposition of ammonium-N (in relation to the average value for the forest interior: 250-1200m, in %) and pH-H_2O of the upper 10 cm of the mineral soil along the transect.

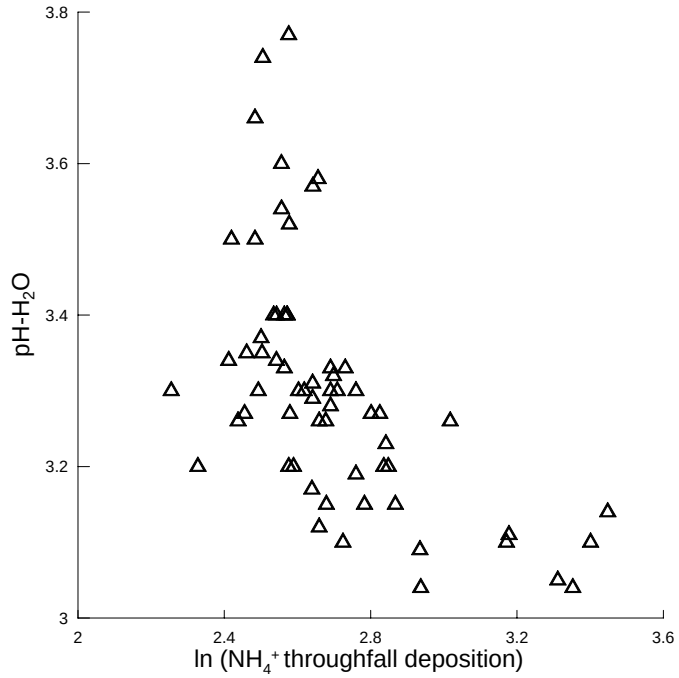

Fig. 3.5: Relation between pH-H_2O of the topsoil and its ln-transformed NH_4^+ throughfall deposition

3.5 Discussion

It is often very difficult to isolate the role of anthropogenic versus natural sources of acidity in affecting processes of soil acidification (Van Breemen et al. 1984). In this case study, the significant range in magnitude of ammonium throughfall depositions (and probably also of sulphate and nitrate; see Fig. 3.2) offered the opportunity to study its impact on soil acidity under comparable site and climatic conditions. We can assume that acidification as a consequence of organic acid deprotonation and forest growth is quite homogeneous along the transect.

We found a significant negative correlation between soil pH and ammonium throughfall deposition. Also Bergholm et al. (2003) found a pH decrease after an experimental addition of ammonium sulphate. NH_4^+ can play an important role as potentially acidifying component, not only because of its high throughfall deposition flux in itself (see also §2.5.3), but also by enhancing the deposition of sulphate and nitrate due to the so-called process of co-deposition (Van Breemen et al. 1982). Signs of co-deposition of SO_2 and NH_3 have also been reported in studies in which fumigation with NH_3 has enhanced SO_4^{2-} concentrations in throughfall (Cape et al. 1995, 1998). NH_3 can pass directly into the needles via the stomata, as well as be retained in the film of water present on the needles or leaves during and after rainfall episodes (Derome et al. 2004). The dissolution of NH_3 in a water film results in the formation of OH^--ions through the reaction (Equation 3.1)

$$NH_3 + H_2O \leftrightarrow NH_4^+ + OH^- \qquad (3.1)$$

and a corresponding increase in pH. If SO_2 gas is present, its dry deposition on the needle surfaces will be promoted by the presence of an alkaline water film on the needle surfaces (Cape et al. 1998), resulting in the formation of ammonium sulphite (Equation 3.2)

$$2NH_4^+ + 2OH^- + SO_2 \leftrightarrow (NH_4)_2SO_3 + H_2O \qquad (3.2)$$

and the subsequent oxidation into ammonium sulphate $(NH_4)_2SO_4$ (Derome et al. 2004).

For the throughfall water collected in the centrally situated ICP monitoring plot, this codepositon was clearly demonstrated by the high and significant correlation coefficients between NH_4^+ and SO_4^{2-}. The strong correlation between NH_4^+ and NO_3^- also suggests a type of co-deposition, although this could not be confirmed by literature data.

The amount of ammonium-N throughfall deposition measured along the transect varied between 10 and 31 kg ha^{-1} during the 6-months measuring period. The highest NH_4^+

throughfall depositions were found over a distance of 270m located close to the forest edge (Fig. 3.4). Also at the north-eastern border higher but less significant enhancements in NH_x throughfall deposition were recorded compared to the forest interior. Several authors agree with the findings of Draaijers et al. (1998) that dry deposition is elevated in forest edges positioned at the forest site with dominant wind direction, in our case south-western winds (see also Chapter 4). In forest edges oriented to other than the prevailing wind directions, higher deposition is restricted to the first few meters of the forest edge caused by some small turbulent currents at the leeside. Compared to other studies (e.g. Beier and Gundersen 1989, Wuyts et al. submitted, Table 4.1), a less steep decrease in ammonium throughfall deposition with distance from the forest border and a larger depth of the forest edge zone was recorded in the present study. This is due to the fact that (as can be seen in Fig. 3.1) the forest edges in our case study are not perfectly perpendicular to the forest border, but show a slightly increasing distance from the forest edge with increasing distance along the transect. The elevated ammonium throughfall deposition at the NE-border shows less elevated dry deposition fluxes. Also Draaijers et al. (1988) found in forest edge zones located at non-dominant wind directions less large and less elevated dry deposition fluxes.

Acidifying (e.g. H_2SO_4, HNO_3) (Ulrich 1986) and potentially acidifying (NH_3) (Nihlgard 1985) deposition were already early hypothesized to reduce soil pH and base saturation, with other possible detrimental effects as e.g. forest decline as a consequence. Many forest top soils are nowadays, however, already very acid and fall within the aluminium buffer range (see Chapter 2), so changes in pH-H_2O are often difficult to observe, unless the soil pH drops from one buffer range to another (Lesturgez et al. 2006) (see also Fig. 1.10). The recorded pH-H_2O-values of the topsoil were very low over the entire transect, all having a pH-H_2O lower than 3.8. This means that over the entire transect length, the topsoil is in the aluminium/iron or iron buffer range (Ulrich 1991). At both the SW and NE forest border, soils were significantly more acid then in the forest interior, and have pH values indicating that soils already moved on to the iron buffer range (pH<3.2).

The acid neutralizing capacity (ANC) of the soil, i.e. the capacity of a soil to buffer or neutralize incoming protons, is provided by various mechanisms (see also Table 1.5): exchange of so-called base cations for protons, adsorption of SO_4^{2-} on Al and Fe-hydroxides for OH^-, dissolution of Al^{3+} from organic complexes and secondary Al minerals, and by silicate weathering. Silicate weathering is the only process that occurs on the whole pH scale. For soil samples taken in the ICP forest monitoring plot at Ravels (see Fig. 3.1), Van Ranst et

al. (2002) calculated the acid neutralizing capacity related to the most important neutralizing substances, i.e. lattice so called base cations in silicate minerals, organic matter (OM) and aluminium. They did find that the acid neutralizing capacity is predominantly related to the silicates present (and not to OM), and the ANC related to so-called base cations is only a fraction of the ANC related to aluminium (Van Ranst et al. 2002).

Table 3.2: Acid neutralizing capacity related to lattice so-called base cations (K^+, Ca^{2+}, Mg^{2+}, Na^+) in silicate minerals (LBC) and aluminium (Al) and organic matter (OM) (in $kmol.ha^{-1}$) in the ICP forest plot at Ravels (Source: Van Ranst et al. 2002)

	LBC	**Al**	**OM**
Forest floor	5.5	57.6	4.2
Mineral top soil	256.3	887.5	3.6

Neutralization of the potentially acidifying input by throughfall water makes that Al^{3+} comes in solution (Cronan and Grigal 1995) leading to increased aluminium toxicity and nutrient imbalances (Van Dijk et al. 1989, Tomlinson, 1991) and aluminization of organic complexes in the forest floor (Van Ranst et al. 2002). The acidification process also leads to a further breakdown of the silicates still present (Van Ranst et al. 2002). In the zones in the forest where soil pH is situated in the iron buffer range, proton buffering may cease since this is accompanied by a shift in the exchangeable cation composition from Al^{3+} to H^+ and Fe^{2+} ions (Ulrich 1991, see also Table 1.5). Deeper soil layers were found to be less acid (numbers not shown), and will probably neutralize seeping H^+ ions.

3.6 Conclusions

From this chapter we can conclude that atmospheric potentially acidifying deposition can be an important cause for soil acidification. In the zones along the transect were NH_4^+ (and probably also NO_3^- and SO_4^{2-}) throughfall deposition is highest, lowest pH-H_2O values were found. At forest edges, atmospheric potentially acidifying deposition can be significantly elevated compared to the forest interior, what can have tremendous consequences for soil pH and Al toxicity to plant roots. Our results indicate that soils can even drop back into the iron

buffer range, meaning that proton buffering ceases and Fe^{2+} ions attain measurable concentrations in the soil solution.

4 On the importance of incorporating forest edge deposition for evaluating exceedance of critical loads

After: De Schrijver A., Devlaeminck R., Mertens J., Wuyts K., Hermy M., Verheyen K. On the importance of incorporation forest edge deposition for evaluating exceedance of critical loads. Applied vegetation science, in press.

4.1 Abstract

A critical load defines the tolerance of natural and semi-natural habitats for anthropogenic air pollution. A correct evaluation of the exceedance of critical loads is therefore fundamental to move towards the long-term protection of ecosystems by limiting emissions of potentially acidifying and eutrophying pollutants. For forest ecosystems, the exceedance of critical loads is often calculated using deposition data measured in the forest interior. However, several studies report forest edges to act as hotspots of potentially acidifying and nitrogen deposition, showing up to four-fold increase in atmospheric deposition compared to the forest interior.

This chapter estimates the relevance of considering the higher deposition load in forest edges for calculating exceedance of critical loads for nitrogen and potentially acidifying deposition. We conclude that if measures to control and reduce atmospheric deposition are based on average deposition fluxes within forest stands, deposition reductions will not be enough for preventing adverse effects. Especially in highly fragmented regions, such as Flanders, this is an issue of high relevance.

4.2 Introduction

Several studies report elevated deposition in forest edges because of local advection, enhanced turbulent exchange and inflow. In fact, forest edges have been shown to act as 'hotspots' for potentially acidifying and nitrogen deposition, showing up to four-fold increase in the rate of atmospheric delivery compared with nearby areas without edges (Weathers et al. 2001). Small forest patches, especially in highly fragmented landscapes, are characterised by a high edge to interior ratio.

Even-though edge enhancement of deposition is substantial, calculations of exceedance of critical loads of nitrogen and acid in forests never take into account this elevated deposition load. In most studies, exceedance of critical loads is calculated using deposition data of the forest interior, causing significant underestimation of ecosystem damage. The concept of critical load (CL) was defined to express the tolerance of natural and semi-natural habitats for anthropogenic air pollution (Nillson and Grennfelt 1988). It relates to the maximum exposure to one or more pollutants without occurrence of any significant harmful effect on for example biodiversity, forest vitality or nitrate seepage to groundwater, according to present knowledge. Two aims of the European directive 2001/81/EC confirm the need for correct calculation of exceedances. The first aim is 'limiting emissions of acidifying and eutrophying pollutants in order to improve the protection of the environment and human health against risks of adverse effects'. The second is 'to move towards the long-term objective not exceeding critical levels and loads and of effective protection of all people against recognized health risks from air pollution'.

This chapter estimates the relevance of considering the higher deposition load in forest edges for calculating exceedance of critical loads for nitrogen and potentially acidifying deposition. We consider Flanders, northern Belgium, as a case study. Average potentially acidifying deposition is among the highest in Europe (UNECE-EC 2003) and about 60% of all forest complexes are smaller than 10 ha, indicating considerable fragmentation, and consequently, a high forest edge/forest interior ratio (Fig. 4.1). This chapter does not offer an exact assessment of the error on the current calculations, but only argues for the relevance of taking into account variability in depositions caused by forest edge effects. We used data of the forest mapping for Flanders and deposition fluxes of six experimental plots of the ICP Forest Monitoring Programme (International Co-operative Programme on assessment and Monitoring of Air Pollution effects on Forests) (Fig. 4.1), which were selected to be representative for the Flemish forest area.

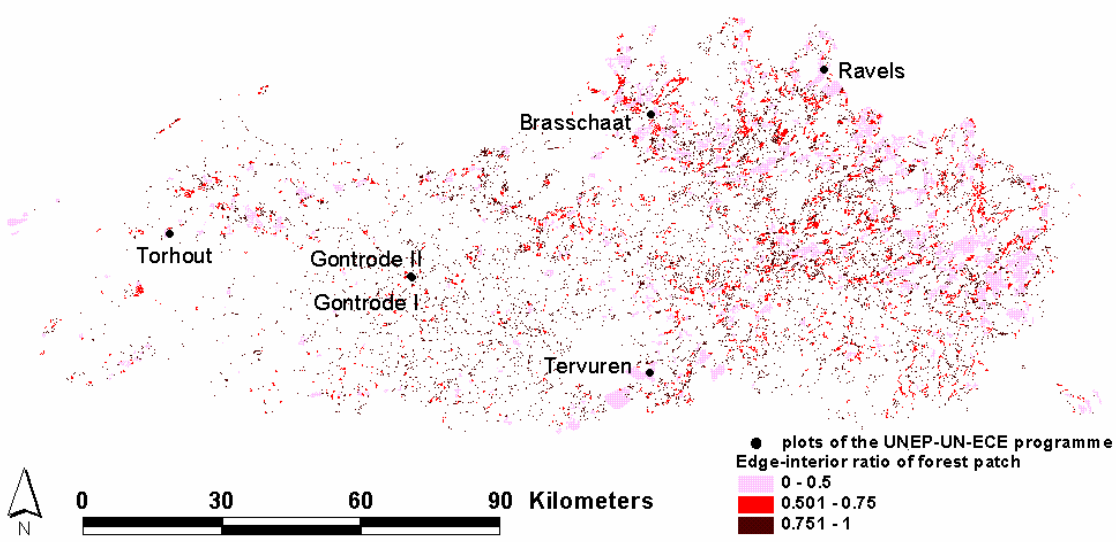

Fig. 4.1: Forest area in Flanders with indication of the edge to interior ratio of different forest patches and the location of the six experimental plots of the ICP Forest Monitoring Program

4.3 Materials and methods

The term 'forest edge distance' was used for the depth-of-influence into the forest under influence of higher stand deposition (sum of throughfall and stemflow deposition). The term 'forest edge effect' defines this increase of stand deposition in the forest edge compared to the forest interior and is usually calculated as the ratio 'average stand deposition in the forest edge / average stand deposition in the forest interior'. Stand deposition is considered to be a good measure of atmospheric deposition input because it is the net result of the interaction between atmosphere and forest canopy.

A literature survey was performed to obtain median values for forest edge distances and forest edge effects (Table 4.1). Hereby eight published studies dealing with 23 case studies investigating atmospheric throughfall deposition in forest edges were considered. This literature survey is not intended to be complete but is rather meant as an indication of the variability on forest edge distance and effect.

The percentage of Flemish forest area under the influence of edge effects was calculated using forest inventory data (MVG *et al*. 2001). Using an average penetration depth obtained from the literature survey, which showed a variation of 15 to 108m (Table 4.1), an edge zone was calculated for each forest patch using ArcView 3.2 (ESRI 2000). Only external forest edges

between forest complexes and non-forested areas were considered. By summing these individual external forest edge areas, the total forest edge area for Flanders was calculated and subsequently expressed as a percentage of total forest area.

Average stand deposition values of inorganic nitrogen (NH_4^+-N + NO_3^--N) and potentially acid ($NH_4^+ + NO_3^- + SO_4^{2-}$) to the forest interior habitat were calculated by averaging 2002 values of forest stand deposition of six Flemish forests of the ICP Forest Monitoring Programme (Genouw et al. 2003). Stand deposition to forest edge habitat was calculated by multiplying the average deposition values of NH_4^+, NO_3^- and SO_4^{2-} for the forest interior with average values of their respective average forest edge effects obtained from the literature survey. Calculation of average deposition values for nitrogen and acid including forest edge effects was done by Equation 4.1:

$$\% FI_{area} \text{ x } FI_{dep} + \% FE_{area} \text{ x } FE_{dep} \qquad (4.1)$$

with FI_{area} =percentage of forest interior area in Flanders,

$FI_{dep.}$ = average stand (throughfall + stemflow) deposition values in the forest interior,

FE_{area} = percentage of external forest edge area in Flanders,

$FE_{dep.}$ = calculated average stand (throughfall + stemflow) deposition values in the external forest edges.

Comparing these average deposition values inclusive of forest edge effect, with average interior deposition values, reveals the underestimation of deposition when edge effects are not included. Comparing both values with the calculated critical loads for Flanders yields the average exceedance of critical loads for the Flemish forest area inclusive and exclusive of external forest edge effects.

Table 4.1: Literature overview of forest edge distance (FED) and forest edge effect (FEE)

		Country	Region	Forest type	FED (*m*)	FEE		
						SO_4^{2-}	NO_3^-	NH_4^+
1	Beier and Gundersen (1989)	Denmark	50 km N Copenhagen	*Picea abies*	15	2.60	4.00	2.80
2	Devlaeminck et al. (2005)	Belgium	Flanders	*Fagus sylvatica*	50	/	/	1.22
3	Draaijers et al. (1988)	The Netherlands	The Veluwe	*Pseudotsuga menziessi*	100	1.80	1.50	1.50
4	Draaijers et al. (1988)	The Netherlands	The Veluwe	*Pseudotsuga menziessi*	50	1.30	1.30	1.20
5	Draaijers et al. (1993)	The Netherlands	Utrechtse Heuvelrug	*Larix decidua*	108	1.00	1.00	1.00
6	Draaijers et al. (1993)	The Netherlands	Utrechtse Heuvelrug	*Pinus sylvestris*	63	1.86	1.43	1.63
7	Draaijers et al. (1993)	The Netherlands	Utrechtse Heuvelrug	*Pinus nigra*	43	5.18	3.83	3.78
8	Draaijers et al. (1993)	The Netherlands	Utrechtse Heuvelrug	*Pinus sylvestris*	65	1.00	1.00	1.00
9	Draaijers et al. (1993)	The Netherlands	Utrechtse Heuvelrug	*Larix decidua*	69	1.98	2.42	1.63
10	Draaijers et al. (1993)	The Netherlands	Utrechtse Heuvelrug	*Pinus abies*	49	4.53	2.41	3.89
11	Draaijers et al. (1993)	The Netherlands	Utrechtse Heuvelrug	*Larix decidua*	77	1.00	1.76	1.00
12	Draaijers et al. (1993)	The Netherlands	Utrechtse Heuvelrug	*Pinus sylvestris*	65	1.00	1.00	1.00
13	Hasselrot and Grennfelt (1987)	Sweden	West coast	*Pinus sylvestris*	50	1.5	2.9	2.7
14	Neal et al. (1994)	England	Hampshire	*Fagus sylvatica*	50			1.5-2?
15	Spangenberg and Kölling (2004)	Germany	Southern Bavaria	*Picea abies*	50-100*	1.00	2.20	1.50
16	Spangenberg and Kölling (2004)	Germany	Southern Bavaria	*Picea abies*	50-100*	1.20	1.80	1.40
17	Spangenberg and Kölling (2004)	Germany	Southern Bavaria	*Picea abies*	50-100*	1.70	1.60	0.70
18	Spangenberg and Kölling (2004)	Germany	Southern Bavaria	*Picea abies*	50-100*	0.60	1.20	1.10
19	Spangenberg and Kölling (2004)	Germany	Southern Bavaria	*Picea abies*	50-100*	0.90	0.60	0.50
20	Spangenberg and Kölling (2004)	Germany	Southern Bavaria	*Picea abies*	50-100*	1.50	2.60	1.80
21	Spangenberg and Kölling (2004)	Germany	Southern Bavaria	*Picea abies/ Fagus sylvatica*	50-100*	0.70	2.20	1.70
22	Weathers et al. (2001)	USA	New York State	Mixed deciduous	25-28	1.12	1.43	1.27
23	Weathers et al. (2001)	USA	New York State	Mixed deciduous	25-28	1.12	1.43	1.27
	Median				50	1.21	1.60	1.50
	Minimum					0.60	0.60	0.50
	Maximum					5.18	3.83	3.78

* To calculate the median FED-value, a FED-value of 50 m was taken for the study of Spangenberg and Kolling (2004) because spatial resolution of sampling was very high.
Presented enhancement factors are calculated as the ratio of the throughfall or net-throughfall (throughfall – bulk deposition) flux
(1) under the first row of trees and the flux at 50 m distance from the forest edge: 1, 2, 13, 15, 16, 17, 18, 19, 20, and 21- (2) at 10 and 200 m from the forest edge: 3, 4- (3) at edge and >25 m from the forest edge: 22-23 (4) at edge and 180 m from the forest edge: 2 and (4) at distance from edge/edge height = 0.25 and at distance from edge/edge height = 5: 4, 6, 7, 8, 9, 10, 11, 12

4.4 Results and discussion

There is substantial discrepancy among recent studies about the intensity and generality of external edge deposition effects (Table 4.1). Temporal as well as spatial variations in ambient air concentrations of pollutants lead to different deposition loads to forest ecosystems (Ivens 1990). Furthermore forest stands differ in their efficiency of intercepting air pollution. This depends upon the prevailing meteorological conditions, stand density, tree height, tree species, management, edge aspect and slope or landscape matrix (Augusto et al. 2002, Erisman et al. 2003). Also differences in methodology and period of sampling may influence the results of deposition studies (Bleeker et al. 2003). Because of this high variability and the synthetic character of this chapter, median literature values of both 'forest edge distance' and 'forest edge effect' were calculated (Table 4.1). Median forest edge distance was 50 m and median forest edge effect was 1.50, 1.60 and 1.21 for NH_4^+, NO_3^- and SO_4^{2-}, respectively.

Considering a depth-of-influence of external forest edge effects of 50 m, it was calculated that 58% of the total forested area in Flanders consisted of external forest edges bordering a non-forested area. This calculation gives only a conservative assessment of edges because it does not consider the effect of internal edges situated within forest complexes such as those between forest stands of different heights or tree density and small roads and canopy gaps. These internal edges are also expected to cause edge effects (Draaijers 1993). It must be noted though, that we did not correct for lower deposition fluxes at forest edges not exposed to the dominant winds, such as those facing southwest. Nor did we account for spatial variations in throughfall fluxes caused by either spatially changing aerodynamic properties of the forest or spatially changing air pollutant concentrations.

Potentially acidifying and nitrogen (throughfall + stemflow) deposition in 2002 varied in the six plots of the ICP Forest Monitoring Programme from 2.84 to 4.08 keq.ha^{-1} and 1.32 to 2.51 keq.ha^{-1} respectively (Table 4.2). These values of stand deposition are considered to be representative for the Flemish forests and are measured beyond the forest edge zone.

Then we calculated the average stand deposition in the forest edge by multiplying the average deposition values of NH_4^+, NO_3^- and SO_4^{2-} for the forest interior with the obtained average values of their respective average forest edge effects (Table 4.1). Calculated average stand deposition in external forest edges amounts to 1.86 (± 0.26) keq.ha^{-1}.yr^{-1} for SO_4^{2-}, 0.98 (± 0.15) keq.ha^{-1}.yr^{-1} for NO_3^- and 2.03 (± 0.60) keq.ha^{-1}.yr^{-1} for NH_4^+. These calculations result

in 4.87 (± 0.99) keq.ha^{-1}.yr^{-1} potentially acidifying deposition and 3.01 (± 0.62) keq.ha^{-1}.yr^{-1} nitrogen deposition.

Table 4.2: Stand deposition (throughfall + stemflow) in 2002 in six plots of the ICP Forest Monitoring Programme of northern Belgium

Plot	**Tree species**	**SO_4^{2-}**	**NO_3^-**	**NH_4^+**	**Sum acid**	**Sum N**
		keq.ha^{-1}	*keq.ha^{-1}*	*keq.ha^{-1}*	*keq.ha^{-1}*	*keq.ha^{-1}*
Torhout	*Fagus sylvatica*	1.36	0.57	1.41	3.34	1.98
Ravels	*Pinus nigra*	1.45	0.66	1.85	3.96	2.51
Brasschaat	*Pinus sylvestris*	1.68	0.76	1.63	4.08	2.39
Gontrode I	*Fagus sylvatica/Quercus robur*	1.60	0.56	1.48	3.63	2.04
Tervuren	*Fagus sylvatica*	1.29	0.64	0.91	2.84	1.55
Gontrode II	*Fraxinus excelsior*	1.85	0.49	0.83	3.17	1.32
Average		1.54	0.61	1.35	3.50	1.96

Source: Genouw et al. 2003

The calculated elevated stand deposition fluxes in external edges, or 58% of the Flemish forest, were corporated in the calculation of average stand deposition fluxes using formula 1: 1.73 keq.ha^{-1}.yr^{-1} for SO_4^{2-}, 0.83 keq.ha^{-1}.yr^{-1} for NO_3^- and 1.74 keq.ha^{-1}.yr^{-1} for NH_4^+, which resulted in 4.30 (± 0.61) keq.ha^{-1}.yr^{-1} for potentially acidifying deposition and 2.57 (± 0.41) keq.ha^{-1}.yr^{-1} for nitrogen deposition.

The median critical load for potentially acidifying deposition to protect forest soils from root damage is 2.75 and 3.09 keq.ha^{-1}.yr^{-1} in coniferous and deciduous forests respectively (Van Avermaet *et al*. 2006, see also Table 1.3). The calculated median critical load value for nitrogen to (1) protect biodiversity is 0.74 and 1.05 keq.ha^{-1}.yr^{-1} in coniferous and deciduous forests respectively and (2) to protect groundwater from nitrate pollution (50 mg.l^{-1} criterion) is 2.90 and 4.90 keq.ha^{-1}.yr^{-1} in coniferous and deciduous forests respectively (Langouche *et al*. 2002, see also Table 1.1). Critical load values for coniferous and broadleaved forests were averaged using the surface area of both forest types (56% deciduous or mixed deciduous forest, 44% coniferous or mixed coniferous forest) as a weight factor, resulting in 2.90 keq.ha^{-1}.yr^{-1} for protecting roots, 0.912 keq.ha^{-1}.yr^{-1} for protecting biodiversity and 4.02 keq.ha^{-1}.yr^{-1} for protecting groundwater from nitrate pollution.

The average exceedance of the critical load for protecting forests from root damage was about 23% higher (0.79 keq.ha^{-1}.yr^{-1}) when elevated deposition fluxes in forest edges were considered (Fig. 4.2). Considering the critical load for protecting biodiversity from nitrogen deposition, it was calculated that the exceedance values were underestimated by 0.61 keq.ha^{-1}.yr^{-1} (31%). The critical load value for protection of groundwater was not exceeded in both cases. These calculations should not be interpreted as exact assessments of the error on the current calculations, but are a clear indication of the need to take forest edge effect into account.

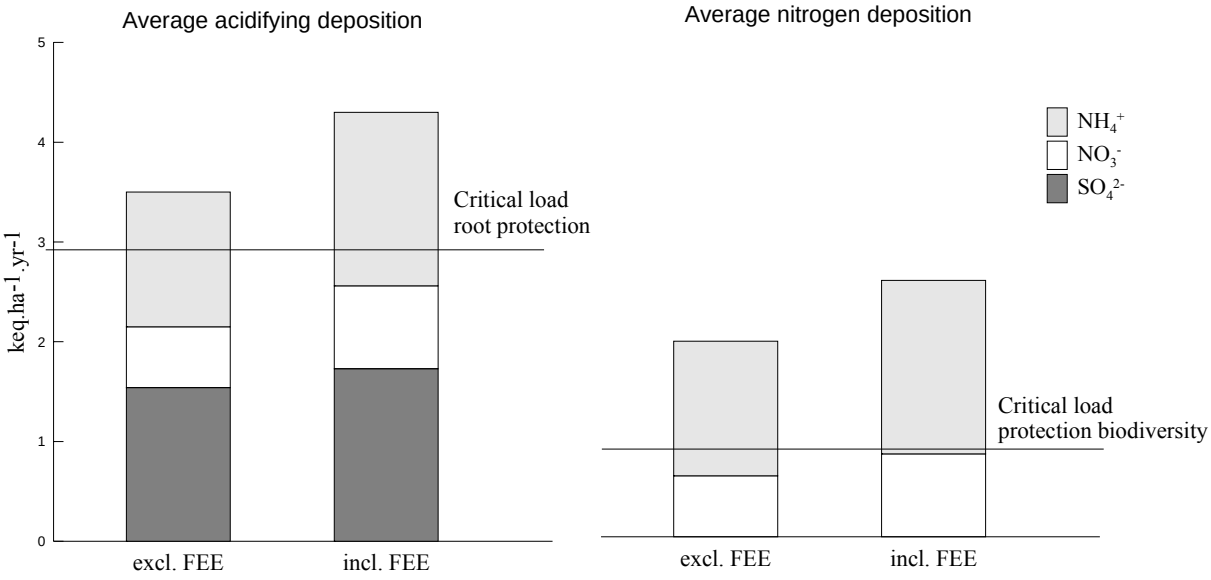

Fig. 4.2: Average stand (throughfall + stemflow) deposition and exceedances of critical load values (in keq.ha^{-1}.yr^{-1}) exclusive (excl.) and inclusive (incl.) forest edge effect (FEE)

If measures to control and reduce atmospheric deposition are based on average deposition fluxes within forest stands, deposition reductions will not be enough for preventing adverse effects. We thus conclude that there is an urgent need to rephrase the calculation of exceedance of critical loads, taking into account edge enhancement of deposition. This is an issue of high relevance, especially in highly fragmented regions, such as Flanders. For exact quantifications of the error on the current calculations, further research is certainly needed.

4.5 Conclusions

When exceedances of critical load values are calculated, elevated deposition in forest edges should be taken into account. We assessed, by means of calculated critical load values for Flanders (by Langouche et al. 2002, see also Table 1.1 and Table 1.3) and averaged throughfall + stemflow deposition values in representative forest ecosystems in Flanders (Plots from the ICP Forest Monitoring Programme), the error on current calculations when elevated depositions in forest edges is ignored. The average exceedance of the critical load for protecting forests from root damage was about 23% higher (0.79 $keq.ha^{-1}.yr^{-1}$) when elevated deposition fluxes in forest edges were considered. Considering the critical load value for protecting biodiversity from nitrogen deposition, it was calculated that the exceedance values were underestimated by 0.61 $keq.ha^{-1}.yr^{-1}$ (31%). The critical load value for protection of groundwater was not exceeded in both cases. These calculations should not be interpreted as exact assessments of the error on the current calculations, but are a clear indication of the need to take forest edge effect into account.

5 Vegetation type effect on throughfall deposition and seepage flux in an area with intermediate deposition load

After: De Schrijver A., Staelens J., Van Hoydonck G., Janssen N., Mertens J., Gielis L., Wuyts K.., Geudens G., Augusto L. & Verheyen K. Vegetation type determines throughfall deposition and seepage quantities. Submitted to Environmental Pollution.

5.1 Abstract

This chapter aims to compare different vegetation types (coniferous and deciduous forest and grassed and pure heathland) in terms of input (throughfall deposition) and output (seepage flux), in a region with relatively low emissions from intensive livestock husbandry. Coniferous forest (2 plots *Pinus sylvestris* and 2 plots *Pinus nigra*) receives significantly higher nitrogen and sulphur throughfall deposition than deciduous forest and both types of heathland. Grassed and pure heathland deliver significantly higher throughfall quantities of Ca^{2+} and Mg^{2+} respectively. The clear differentiation between the vegetation types for throughfall deposition was not univocally reflected in the global clustering of ion seepage. Considerable seepage fluxes of NO_3^-, SO_4^{2-}, Ca^{2+} and Al(III) were only found under the *Pinus nigra* plots. We discuss our hypothesis that the *Pinus nigra* stands already have evolved to a situation of nitrogen saturation, while the other vegetation types have not.

5.2 Introduction

Many terrestrial ecosystems in Europe and some parts of North America have evolved from nitrogen limited to nitrogen saturated during the past decades (Gundersen et al. 2006). Semi-natural ecosystems originally adapted to nutrient poor conditions and using nutrients sparingly, nowadays experience excess of nutrients and therefore evolve towards vegetation types with more nutrient-demanding species (Smart et al. 2005). For example heathland ecosystems' characteristics have changed dramatically and grasses (*Molinia caerulea* or *Deschampsia flexuosa*) have replaced heather completely as a dominant species in many heathlands (Aerts et al. 1990). The evolution to ecosystems in which the availability of inorganic nitrogen is in excess of total combined plant and microbial nutritional demand (Aber et al. 1989), is a pernicious consequence of chronic nitrogen additions. Within the next decades, considerable increases in nitrogen emissions are predicted in other parts of the word (e.g. Asia) (Galloway et al. 2004), so nitrogen saturation of ecosystems may become a world-wide problem. Furthermore, recent changes in global and regional climate may accelerate this process via enhanced mineralization of soil N (Gundersen et al. 2006).

In nitrogen limited ecosystems, biogeochemical cycling of nitrogen is an almost closed internal cycle mainly driven by litter production and quality, decomposition, mineralization, immobilization and plant uptake (Gundersen et al. 2006). In regions with high atmospheric pollution load, the capacity of an ecosystem for scavenging atmospheric pollution plays an additional role in its biogeochemistry and certainly partly determines the time of onset of nitrogen saturation. Within the great variety of forest ecosystems, tree species composition (Augusto et al. 2002), forest age and the accompanied leaf area index and forest height (Erisman and Draaijers 2003) are reported to be driving factors for dry deposition.

The nowadays experimental evidence and datasets of throughfall deposition and seepage quantities in (semi-)natural ecosystems are dominated by coniferous forest (Gundersen et al. 2006). Less is known about deciduous forest ecosystems and only limited comparisons are made between different forest types in similar site and climatic conditions (De Schrijver et al. in press). Moreover, comparisons of forest with other vegetation types, e.g. heathland, are rarely existing (e.g. Herrmann et al. 2005). For model predictions and simulations of nitrogen fluxes at the catchment scale, it is necessary to gain insight whether vegetation type influences the throughfall input and seepage output of ions. This chapter therefore aims to test the hypothesis that vegetation types differ in their throughfall deposition and seepage fluxes. We compare data of throughfall deposition and seepage fluxes in coniferous and deciduous forest

and two types of heathland ecosystems. The experiments were performed in a region with intermediate nitrogen load (<20 kg $N.ha^{-1}.yr^{-1}$ via bulk deposition), at sites with comparable site and climatic conditions.

5.3 Materials and methods

5.3.1 Site description

The study area was situated in Flanders (northern Belgium), in the nature reserve 'Vallei van de Zwarte beek' (54°03' N, 05°16'E), which covers an area of 1500 ha, including forest (especially homogeneous plantations of *Pinus nigra ssp. laricio* Maire and *Pinus sylvestris* L.), marshland, grassland, heathland and pasture land.

The experiments were carried out in four different deciduous forest sites with dominance of *Quercus robur* L. and/or *Betula pendula* Roth and in four different coniferous forest sites of which two stands of *Pinus sylvestris* and two stands of *Pinus nigra ssp. laricio* (Table 1.1). In a representative circular plot of 500 m^2, all trees were measured for their diameter on breast height and total tree height. These parameters were used to calculate the plot average basal area (m^2) and stand volume (m^3) (Table 1.1) by means of the equations reported by Dagnelie et al. (1985).

Table 5.1: Stand characteristics of the forested plots (N=stem number, G=basal area on breast height, V=total stand volume, D=average stem diameter at breast height and H=average tree height)

Plot Code	**Vegetation type**	**N**	**G**	**V**	**D**	**H**
			(m^2/ha)	*(m^3/ha)*	*(m)*	*(m)*
D1	*Quercus/Betula*	300 (180/120)	17	123 (73/50)	0.29	13
D2	*Quercus/Betula*	620 (179/421)	26	214 (114/99)	0.29	16
D3	*Quercus/Betula*	1180 (936/24)	18	108 (86/16)	0.13	10
D4	*Quercus*	500	17	125	0.19	12
Ps1	*Pinus sylvestris*	840	41	351	0.25	19
Ps2	*Pinus sylvestris*	500	38	230	0.26	18
Pn1	*Pinus nigra*	500	51	539	0.35	23
Pn2	*Pinus nigra*	420	44	457	0.36	23

Furthermore, four plots were selected in heathland vegetation, of which two (H1 and H2) with >80% cover of *Calluna vulgaris* L. Hull, and two (GH1 and GH2) with >80% cover of *Molinia caerulea* L. Moench. In H1 and GH2, *Erica tetralix* occurs in small covers.

The history of the selected stands was reconstructed using historical topographic maps (Fig. 5.1). The coniferous forest plots were transformed from heath to pine in the period 1853-1939. Three deciduous plots were conversions of pine or mixed stands (natural development in D1 and D3, and planted in D4), while plot D2 was planted between 1950 and 1971 after a short period of heathland vegetation. The heathland plots were all covered by heath at least since 1771. All plots were sandy podzols with pH-KCl-values of the forest floor ranging from 2.8 to 3.5 (Table 5.2) and C/N ratios from 18 to 36. The average year-round groundwater level, measured in piezometers, was lower than 1.8 m for all forest plots. In the heathland plots, average groundwater level ranged between 0.4 and 0.8 m (Table 5.2).

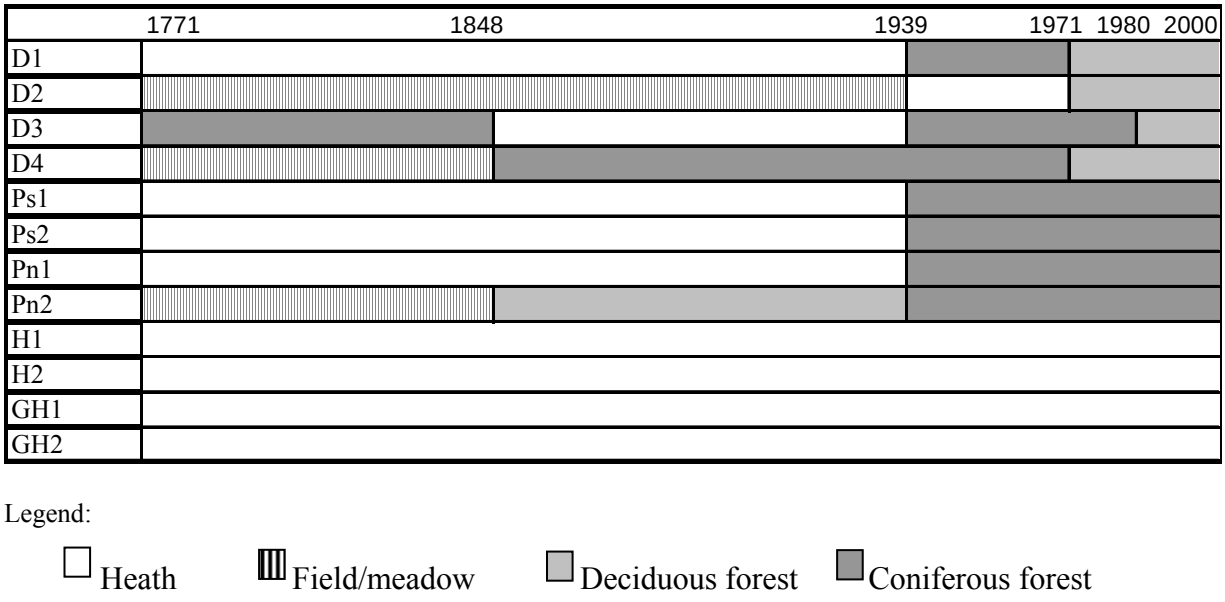

Fig. 5.1: Overview of historical land use of the study plots

Table 5.2: pH-KCl, C/N values of the forest floor and the upper mineral soil (0-10cm) and mean groundwater level (GWL)

	pH		**C/N**		**GWL**
	Forest floor	Mineral soil	Forest floor	Mineral soil	*(m)*
D1	3.1	3.5	23	38	4.9
D2	3.0	3.2	18	25	2.4
D3	3.2	3.4	25	30	2.4
D4	3.0	3.2	22	24	>5m
Ps1	2.8	3.2	28	33	>5m
Ps2	2.8	3.1	24	29	1.8
Pn1	2.9	3.3	24	24	2.0
Pn2	2.8	3.2	22	18	>5m
H1	3.4	3.7	30	56	0.4
H2	3.5	3.6	31	42	0.8
GH1	3.2	3.4	36	52	0.8
GH2	3.2	3.5	32	29	0.5

5.3.2 Experimental set-up

To avoid a forest edge effect on the deposition loads (Draaijers et al. 1988, De Schrijver et al. in press, Chapter 4), all forest plots were located at least 50 m from the forest edge. Each plot was equipped with ten throughfall collectors and three suction lysimeters with ceramic cups. The porous cup lysimeters were placed beneath the rooting zone (1 m under forest and 0.25 m under heathland) and a –50 kPa suction was applied. Bulk deposition was collected at three locations using four bulk collectors placed above the heath. Throughfall and bulk precipitation were sampled using polyethylene funnels (15 cm diameter) placed at a standard height of 1 m and supported by tubes draining into two-litre polyethylene bottles. A nylon mesh was placed in the funnel to avoid contamination by large particles. The bottles were placed below ground level to avoid the growth of algae and to keep the samples cool. Stemflow water was not collected because of its low contribution to nutrient fluxes in pine and young birch stands.

Throughfall below heathland canopies was collected using a system with slanting half-open channels with a capturing surface of 165 cm² (Bobbink and Heil 1992a). This type of collector was constructed of four parallel gutters draining in a central gutter perpendicular to it, draining in a bottle. At each connection between the parallel and central gutters, a nylon mesh was placed in the funnel to avoid contamination by large particles.

Water fractions were collected and measured on a monthly basis during one year starting from 15 May 2000. On each sampling occasion, the water volume in every collector was measured in the field, and the bottles were replaced by bottles rinsed with distilled water. The throughfall samples of each plot were pooled to one sample for the chemical analyses. All water samples were transported and stored at a maximum temperature of 5°C. After the samples had been analysed for pH (ion-specific electrode), they were filtered through a glass microfibre filter (WHATMAN GF/A) and a nylon membrane filter (GELMAN, nyloflo) of 0.45 μm. Samples were analysed within a week for Cl^- and NO_3^- (ion chromatography), NH_4^+ (photometric determination of a reaction product of NH_4^+ at λ=660 nm according to the Dutch standard method NEN 6576), and K^+, Ca^{2+}, Mg^{2+}, Na^+ and Al(III) (flame atomic absorption spectrophotometry).

5.3.3 Quality control

The quality of the chemical analyses was checked by including method blanks, repeated measurements of internal and certified reference samples, and by inter-laboratory tests. The methods were validated with samples from inter-laboratory profession tests and standard reference materials (CRM 100). For water samples the coefficient of variation based on repeated measurements of internal quality controls was 4.6% for NH_4^+, 3.3% for NO_3^-, 2.4 for Cl^-, 5.1% for Na^+, 4.3% for K^+, 4.1% for Ca^{2+}, 4.3% for Mg^{2+}, and 4.7% for Al(III).

5.3.4 Data handling and statistical analysis

5.3.4.1 Calculation of throughfall deposition and canopy exchange fluxes

Ion bulk and throughfall deposition was calculated by multiplying the water volume with the ion concentration in that volume. Total inorganic nitrogen via bulk and throughfall deposition was calculated by summing NO_3^--N and NH_4^+-N throughfall deposition. Total potentially acidifying deposition (keq.ha^{-1}.yr^{-1}) was calculated as the difference between potentially acidifying (NO_3^-+NH_4^++SO_4^{2-}+Cl^-) and non-acidifying (Ca^{2+}+Mg^{2+}+K^++Na^+) throughfall deposition (UBA 2004).

Compared to bulk precipitation, the chemical composition of throughfall water is generally altered, and it is widely acknowledged that this transformation results from (1) the washing

off of dry deposition of aerosols and gases, as well as (2) canopy leaching, i.e. release of ions from plant tissues or (3) canopy uptake (Parker 1983).

To estimate the contribution of dry deposition and canopy leaching/uptake to net throughfall deposition, we used the canopy budget method developed by Ulrich (1983d) and extended by De Vries *et al.* (1998). However this model is based on some assumptions that are not secure, it has been frequently used (e.g. Bredemeier 1988, Ivens 1990, Van Ek and Draaijers 1994, Houle et al. 1999, Armbuster 2002, Ukonmaanaho and Starr 2002, Langusch et al. 2003, Oyarzun et al. 2004). Dry deposition and canopy exchange of K^+, Ca^{2+} and Mg^{2+} is within this model calculated with (i) the use of bulk deposition instead of wet-only deposition, (ii) the assumption that K^+, Ca^{2+} and Mg^{2+} containing particles are deposited with equal efficiency as particles containing Na^+ and (iii) the assumed relation between wet and dry deposition of particles. In spite of these assumptions, Draaijers et al. (1998) stated that canopy exchange and dry deposition of K^+, Ca^{2+} and Mg^{2+} can be estimated reasonably well using throughfall and bulk precipitation measurements in association with the canopy budget model. Furthermore, no corrections are made for possible leaching of base cations with e.g. organic acids. The reliability of the calculated ammonium uptake by the canopy (as has been used in Chapter 6 (§6.3.4) depends on (i) the calculated base cation exchange, and (ii) the assumed relationship between canopy leaching of base cations and canopy uptake of ammonium and protons, and has been further discussed in §6.5.1.

Following Parker (1983), we calculated the deposition quantity of net throughfall water (NTW, mol ha^{-1} yr^{-1}) to obtain the total effect of the canopy on deposition in the forest (Equation 5.1)

$$NTW = TF - BD = DD + CL \tag{5.1}$$

where *BD* = bulk deposition, *TF* = throughfall, *DD* = dry deposition, and *CL* = canopy leaching.

In the canopy budget method, Na^+ is assumed to be inert with respect to the canopy, i.e. neither uptake nor leakage occurs. Furthermore, particles containing K^+, Ca^{2+} and Mg^{2+} are assumed to have the same deposition velocity as Na^+, as expressed by a dry deposition factor (DDF) (Equation 5.2).

$$DDF = \frac{(TF - BD)_{Na}}{BD_{Na}} \tag{5.2}$$

Dry deposition of K^+, Ca^{2+} and Mg^{2+} is then calculated as bulk deposition multiplied by this dry deposition factor (Equation 5.3)

$$DD_X = BD_X \cdot DDF \tag{5.3}$$

where x = K^+, Ca^{2+} or Mg^{2+}

Canopy exchange of so-called base cations is calculated by subtracting the estimated dry deposition from net throughfall deposition.

5.3.4.2 Calculation of ion seepage fluxes

Ion seepage flux was calculated by multiplying the calculated water seepage flux at 1 m depth with the average ion concentration in the soil solution at 1m depth. Since meteorological parameters were not measured in the stands, the percolation water flux at 1 m depth was calculated using Na^+ as a tracer. We used the method of chloride mass balance (CMB) (Eriksson and Khunakasem 1969), but with Na^+ instead of Cl^- as a tracer. This method is originally based on the assumption of conservation of mass between the input of atmospheric chloride and the chloride flux in the subsurface (Eriksson and Khunakasem 1969) according to Equation 5.4.

$$S = \frac{TD}{Cl_{ss}} \tag{5.4}$$

where S=water seepage flux, TD=total atmospheric chloride deposition and Cl_{ss}=average chloride concentration in the soil solution.

Since Cl^- was recently found to participate in a complex biogeochemical cycle involving the formation of organically bound chloride (Oberg and Sanden 2005), we used Na^+ as tracer. This method seems to be applicable for the observed sandy soils since their low Na^+ content and assuming that during this short experiment, contribution of weathering to Na^+ concentrations in the soil solution is limited.

To compare the throughfall deposition fluxes of the vegetation types throughout time, a repeated measures test (also called: a within-subjects test) was performed. This is a variant on the Factorial Design (an experiment which tests the effect of one or more factors) in which at least one factor is a repeated measure. In this case, time (within factor) and vegetation type (between factor) are two factors determining throughfall depositions. A repeated measures test is more powerful than a one-way Anova test since the procedure accounts for variability

in time. This information is used to provide a more precise estimate of the experimental error. The repeated measures test was conducted after testing the preconditions of normality and homoscedasticity (Neter et al. 1996). Data were all log-transformed to fulfil both preconditions. The major problem with repeated measures designs is the potential for carry-over effects. Therefore, the precondition of absence of autocorrelation was tested (Neter et al., 1996) by means of the Durbin-Watson test, and found to be satisfactory. Furthermore, to fulfil the condition of sphericity (Mauchly's test), the degrees of freedom of the F test were multiplied by the Greenhouse-Geisser epsilon value and the significance of the F test is evaluated with the new degrees of freedom. These analyses were performed (1) on a total year basis and (2) during the growing (period May to November) and (3) the dormant season (period December to April) separately.

To compare the yearly canopy exchange fluxes between the vegetation types, one-way Anova tests were performed on the log-transformed values. The vegetation types were divided in homogenous groups by means of the Duncan posthoc test. This test was also used to compare the yearly seepage fluxes.

To find out whether the vegetation types show a general differentiation in both throughfall deposition and seepage quantities, the respectively datasets (including all ions) were subjected to a hierarchical cluster analysis (average linkage clustering). The vegetation plots were ordered into groups in a way that the degree of association between two plots is maximal if they belong to the same group and minimal otherwise.

All statistical data analysis was performed using SPSS12.0 for Windows.

5.4 Results

5.4.1 Throughfall deposition and canopy exchange

During the 12-month measuring period, total bulk precipitation amounted to 1070 mm. Average interception by the canopy represented on average 25% of the total precipitation for the deciduous stands, 28% for the stands of *Pinus sylvestris*, 35% for the stands of *Pinus nigra* and 14% and 12% for the heathland and grassed heathland plots.

Average throughfall deposition of NH_4^+ and NO_3^- is in both types of heathland lower than bulk deposition (Table 5.3) and this during both the growing and the dormant season. Average throughfall deposition of sulphur is lower in the heathland plots with *Molinia* (plots GH11

and GH12) than bulk deposition which is most apparent during the growing season. Throughfall deposition of the cations Na^+, K^+, Ca^{2+} and Mg^{2+} and of Cl^- is for all vegetation types higher than bulk deposition.

Comparison of the different vegetation types clearly shows that the coniferous forest types receive the highest amounts of NH_4^+, NO_3^-, SO_4^{2-} and Na^+ via throughfall water. Calculation of total inorganic N and the total potentially acidifying deposition shows highest values for the coniferous plots (Fig. 5.2). For these ions, a clear and significant relationship could be found (R^2=0.83 for NH_4^+, R^2=0.82 for NO_3^-, R^2=0.55 for SO_4^{2-} and R^2=0.71 for Na^+) between the net throughfall deposition quantity and the vegetation height (e.g. for Na^+ in Fig. 5.3). K^+ throughfall deposition has the tendency to be highest in the deciduous stands and Ca^{2+} and Mg^{2+} in the heathland plots. No significant influence of vegetation height was found for these ions.

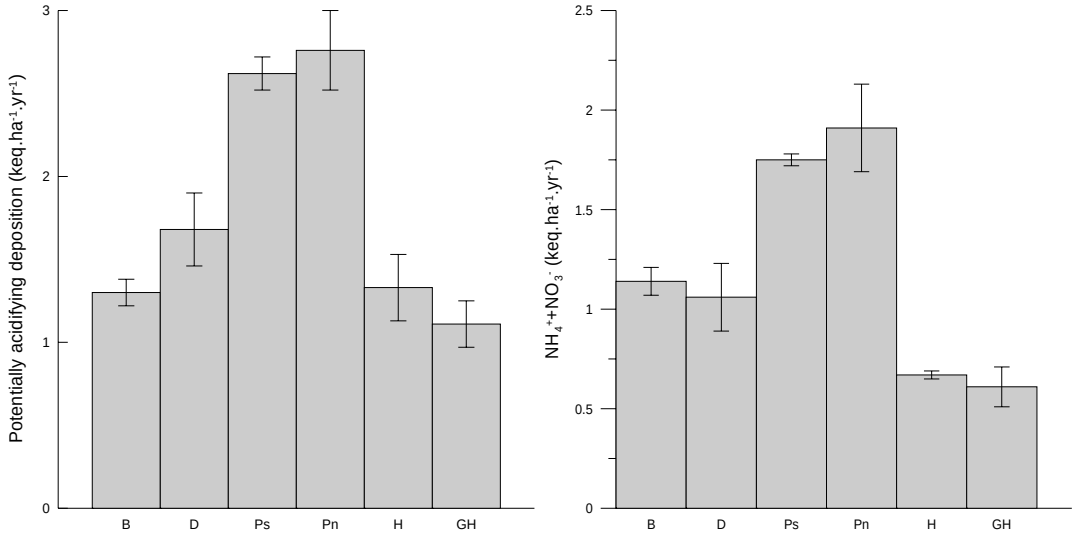

Fig. 5.2: Potentially acidifying and inorganic N bulk (B) and throughfall deposition (keq/ha) (with indication of standard deviation) the different vegetation types (D=deciduous stands, Ps= *Pinus sylvestris*, Pn=*Pinus nigra*, H=heathland, GH=grassed heathland)

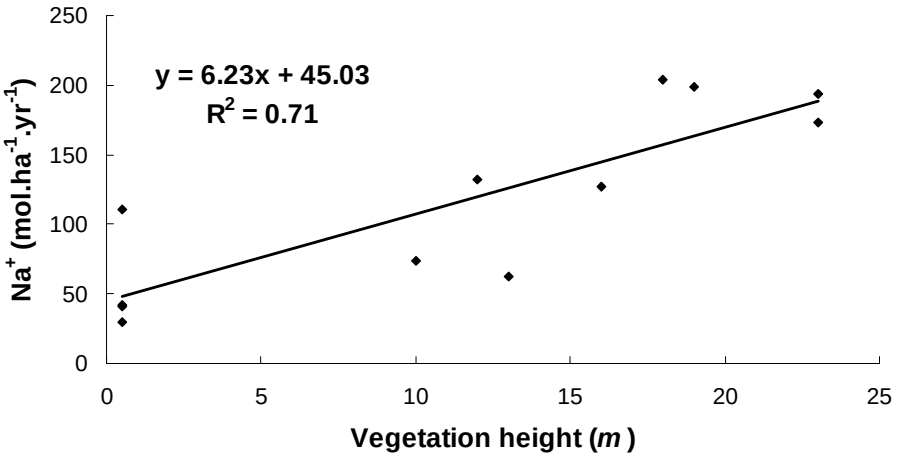

Fig. 5.3: Relationship between vegetation height and Na^+ net throughfall (=throughfall minus bulk) deposition ($mol.ha^{-1}.yr^{-1}$)

Cluster analysis of the total dataset makes distinction between the forest and the heathland plots (Fig. 5.4). Within the forest plots, coniferous and deciduous plots are grouped separately. The coniferous plots are very similar (having the same rescaled distance), while the deciduous plots show higher variability. The heathland plots are divided according to the presence of *Molinia*.

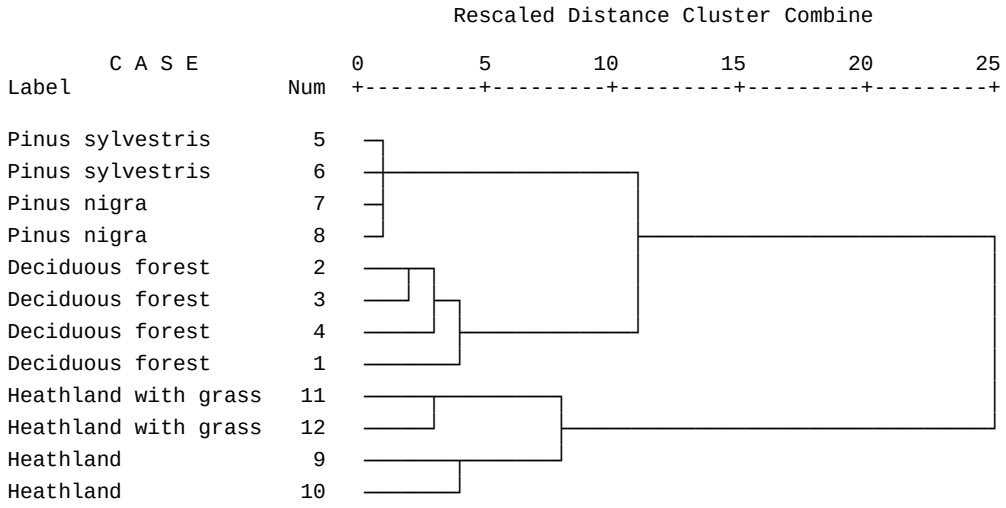

Fig. 5.4: Dendrogram of the hierarchical cluster analysis based on throughfall data of all plots

The outcome of the repeated measures test (Table 5.3) shows that for each considered ion and for the waterflux, time was a significant factor ($p < 0.001$). Only for NO_3^- and for the waterflux, there was a weak interaction between the factors time and vegetation type. For each ion, except for Mg^{2+}, vegetation type was a significant factor. The post-hoc analysis (Table 5.3) shows significant differences in NH_4^+ and NO_3^- throughfall deposition between the heathland plots (plots H and GH), the deciduous forest plots (D) and the coniferous forest plots (Pn and Ps). These results account on a total year basis, as well as during the growing and the dormant season, except for NH_4^+-N during the growing season in which no differences were found between the heathland and the deciduous forest plots.

For SO_4^{2-}, throughfall deposition in the plots of *Pinus sylvestris* and *Pinus nigra* is significantly higher than in the heathland plots with *Molinia*. Na^+ throughfall deposition is significantly higher in both coniferous forest plot types compared to the other vegetation types. Ca^{2+} throughfall deposition is significantly higher in the heathland plots with *Molinia* compared to the other vegetation types. For Mg^{2+}, throughfall deposition is significantly higher in the pure heathland plots than in the stands of *Pinus nigra* except during the growing season. When forest height is included as covariable in the repeated measures test, no appreciable differences were found between the vegetation types.

The outcome of the one-way Anova calculations of canopy exchange of K^+, Ca^{2+} and Mg^{2+} show that in all forest plots, K^+ is the dominant cation exchanged by the canopy, although no significant differences were found between the forest and heathland plots. For Ca^{2+} canopy exchange values of the heathland plots with *Molinia* were found to be significantly higher than for both coniferous forest types, while for Mg^{2+} exchange values were significantly higher in the pure heathland plots than in the coniferous plots (Fig. 5.5).

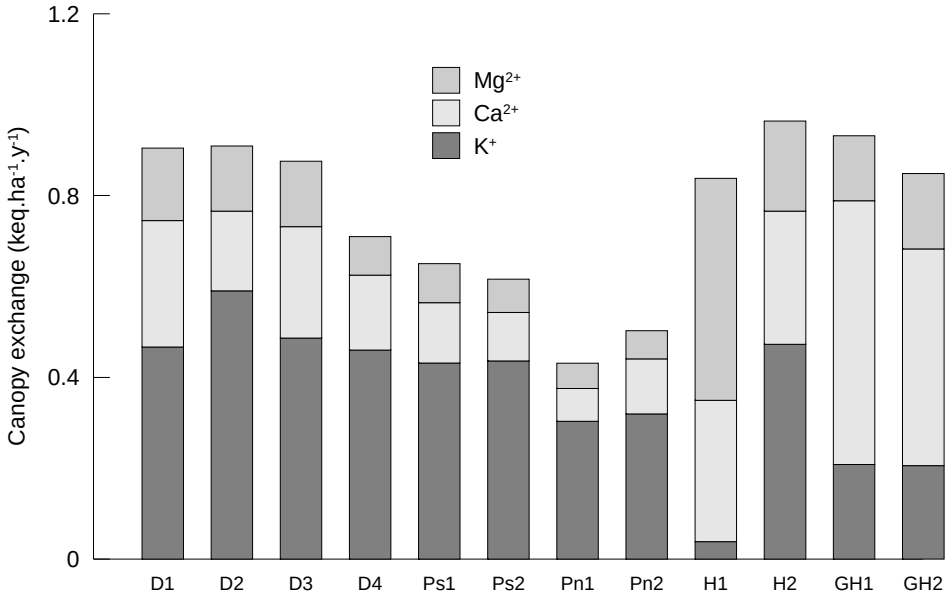

Fig. 5.5: Canopy exchange (keq.ha^{-1}.yr^{-1}) of K^+, Ca^{2+} and Mg^{2+} in the 12 study plots

5.4.2 Seepage flux

Average seepage fluxes of NH_4^+-N were not significantly different between the four vegetation types, and were low compared to NO_3^--N seepage. Significantly (p=0.023) higher NO_3^- seepage fluxes were found under the *Pinus nigra* plots compared to the two types of heathland plots (Table 5.4). Also seepage fluxes of SO_4^{2-} were significantly (p<0.001) highest under *Pinus nigra* and lowest under heathland. K^+ and Mg^{2+} seepage fluxes were not significantly differing under the studied vegetation types, while Ca^{2+} and Al (III) seepage fluxes were significantly (p=0.008 and 0.017) higher under *Pinus nigra* compared to respectively one or both types of heathland (Table 5.4).

Cluster analysis of the seepage dataset (Fig. 5.6) again makes distinction between the forest and heathland plots. Within the forest plots, *Pinus nigra* plots were grouped separately from the other forest plots. Within the heathland plots, distinction is made between two groups which can not be attributed to the vegetation type. Also the further grouping of the forest plots is irrespective of forest type.

Table 5.3: Mean values of waterflux in mm, bulk and throughfall deposition fluxes of different ions (mol.ha^{-1}.yr^{-1}) per vegetation type during the total year, the growing and the dormant season. The groups distinguished by the post-hoc test (Duncan: repeated measures test) are also indicated

Plot	Water	NH^+_4-N	NO^-_3-N	DIN	SO_4^{2-}-S	Na^+	K^+	Ca^{2+}	Mg^{2+}
Total year									
Bulk	1070±54	829±64	471±21	1300±64	269±22	244±4	92±36	115±10	41±4
D-Mean	806±48 ab	639±159 b	427±36 b	1066±306 b	306±29 ab	329±34 a	551±66 b	172±21 ab	92±14 ab
Ps-Mean	767±21 ab	1111±5 c	639±35 c	1750±30 c	434±35 b	433±3 b	514±4 b	166±9 ab	83±4 ab
Pn-Mean	692±5 a	1211±157 c	693±61 c	1904±217 c	428±9 ab	415±15 b	402±7 b	166±12 a	78±0 a
H-Mean	914±17 ab	364±51 a	307±30 a	671±20 a	328±57 ab	307±46 a	445±116 b	203±19 b	115 ± 17 b
GH-Mean	936±19 b	264±30 a	350±71 a	614±101 a	248±20 a	265±6 a	256±25 a	327±67 c	103 ± 6 ab
Growing season									
Bulk	494±35	371±29	243±9	613±39	133±24	122±4	40±16	58±5	16±2
D-Mean	482±28	355±139 a	239±24 b	594±139 b	155±19 ab	157±36 ab	378±64 b	105±18 a	54±9 a
Ps-Mean	486±13	587±5 b	366±33 c	953±38 c	205±10 b	187±4 b	359±21 b	94±1 a	42±1 a
Pn-Mean	439±3a	744±301 b	422±69 c	1166±370 c	222±32 b	201±17 b	234±54 ab	99±16 a	43±6 a
H-Mean	518±9	195±61 a	150±30 a	345±31 a	137±7 ab	132±12 a	264±31 a	100±8 a	55±1 a
GH-Mean	502±14	116±6 a	190±52 a	306±46 a	114±4 a	127±14 a	192±49 a	188±88 b	54±18 a
Dormant season									
Bulk	576±60	458±36	228±9	687±40	136±24	122±8	52±20	58±5	25±3
D-Mean	324±19	282±32 b	188±13 b	470±43 b	152±18 ab	172±31 ab	171±29 ab	67±8 a	39±7 ab
Ps-Mean	281±8	523±9 c	274±1 c	797±10 c	230±26 b	244±1 b	156±18 b	72±9 a	41±4 ab
Pn-Mean	253±2	468±142 c	271±6 c	739±148 c	207±41 b	214±32 b	168±62 b	67±3 a	34±6 a
H-Mean	395±7	170±14 a	160±1 a	329±13 a	191±52 ab	175±36 ab	181±146 b	103±10 b	60±17 b
GH-Mean	435±33	147±34 a	159±18 a	306±52 a	135±24 a	139±7 a	62±23 a	139±20 c	49±13 ab

Table 5.4: Yearly water percolation fluxes (mm) and ion seepage fluxes (mol.ha^{-1}.yr^{-1}) in the 12 study plots

	Water	**NO_3^--N**	**NH_4^+-N**	**DIN**	**SO_4^{2-}-S**	**K^+**	**Ca^{2+}**	**Mg^{2+}**	**Al(III)**
D1	214	79	7	86	250	28	80	33	185
D2	197	186	7	193	324	66	95	53	237
D4	271	200	14	214	318	146	192	103	100
Ps1	207	264	7	271	299	66	145	49	300
Ps2	239	136	7	143	396	56	132	37	208
Pn1	115	636	7	643	661	90	259	53	363
Pn2	172	1414	7	1421	583	61	264	66	582
H1	310	71	21	93	72	118	65	33	167
H2	512	21	7	29	122	69	55	29	126
GH1	436	50	14	64	90	87	77	41	0
GH 2	392	29	14	43	115	41	50	29	93
Mean-D	227±39 ab	155±66 ab	10±4 a	164±69 ab	297±42 a	80±60 a	122±61 ab	63±36 a	174±69 ab
Mean Ps	223±23 ab	200±91 ab	7±0 a	207±91ab	348±68 a	61±7 a	138±9 ab	43±9 a	254±66 ab
Mean Pn	143±40 a	1025±551 a	7±0 a	1032±551 a	622±55 a	75±20 a	262±4 a	60±9 a	473±155 a
Mean H	411±143 b	46±35 b	14±10 a	61±45 b	97±35 b	93±34 a	60±7 b	31±3 a	146±29 ab
Mean GH	414±31 b	39±15 b	14±0 a	54±15 b	103±18 b	64±33 a	64±19 b	35±9 a	46±66 b

Note: For plot D3 no seepage fluxes could be calculated because of the low number of repetitions for soil solution concentrations.

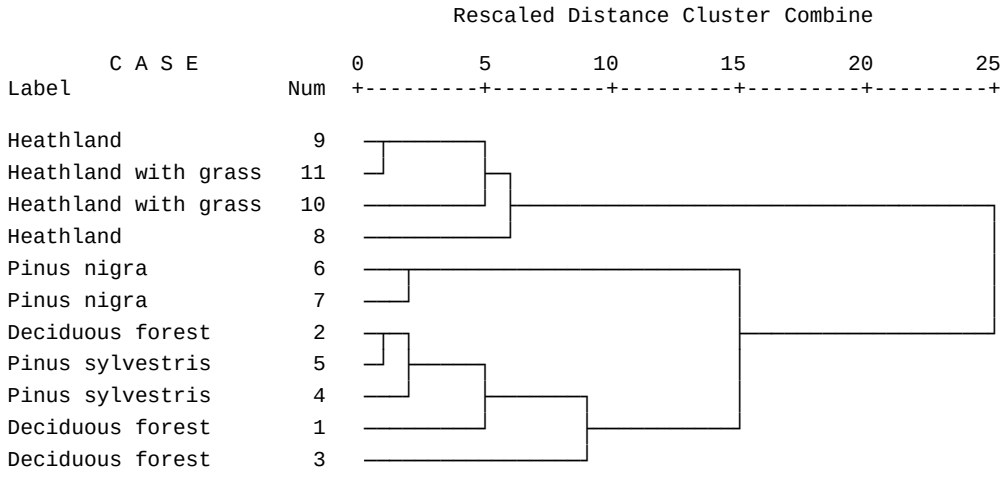

Fig. 5.6: Dendrogram of the hierarchical cluster analysis based on seepage data of all plots.

5.5 Discussion

5.5.1 Do vegetation types differ in their throughfall deposition fluxes?

The studied vegetation types show a general differentiation in throughfall deposition fluxes: both types of heathland, which are separated in two clusters, are clearly distinguished from forest, which in turn is clearly divided in coniferous and broadleaved forest. This clear division in separate clusters shows that each vegetation type has its specific characteristics influencing the input of pollutants/nutrients to the forest floor. Coniferous forests annually, but also during the dormant and growing season separately, receive higher nitrogen and sulphur throughfall deposition than deciduous forest and both types of heathland. The pine plots in this case-study are characterized by the highest trees and stand volume and have evergreen foliage, which are all factors favouring dry deposition (Erisman and Draaijers 2003). We found a significant relationship between the height of the studied vegetation types and the throughfall deposition quantity for ions that are principally dry deposited. Also Fowler et al. (1999) found an increased atmospheric deposition with increasing aerodynamic roughness, resulting in higher deposition rates to forests than to e.g. moorland. In addition, the leaf shape affects the amount of ions deposited. Leaves with long narrow shape (needles) are according to Woodcock (1953 in Smith 1981) more efficient in salt accumulation than circular ones. Differences between the two forest types in throughfall deposition flux are likely to be highest during the dormant season because differences in leaf area index are highest during that period. However, dependent on the observed tree species, its stand

characteristics and the emission regime, throughfall ammonium deposition fluxes can be lower in coniferous forests during the growing season (Cappellato et al. 1993) as well as during the whole year (Cappellato et al. 1993, Houle et al. 1999, Neary and Gizyn 1994). We also discussed this topic in chapter 7.

Part of the dry deposited quantities of NH_4^+ and NO_3^- is assimilated by the leaves and shoots of the heathland and the majority of the deciduous plots. This can be concluded from the lower amounts of NH_4^+ and NO_3^- throughfall deposition compared to its bulk deposition. In the coniferous plots, canopy uptake of NH_4^+ probably also occurs, but this may be obscured by the high dry deposition, with a higher throughfall compared to bulk deposition flux as a result. Uptake of nitrogen out of the atmosphere was previously reported for heath (Bobbink et al. 1992b, Hansen and Nielsen 1998) and for both deciduous and coniferous forest ecosystems (Lovett and Lindberg 1993; Harrison et al. 2000).

Heathland plots are, compared to the other vegetation types, delivering significantly higher throughfall quantities of Ca^{2+} in the case of grassed heathland, and Mg^{2+} in the case of pure heathland. We used the Ulrich (1983d) model for calculating the canopy exchange of NH_4^+ for K^+, Ca^{2+} and/or Mg^{2+} for all observed ecosystems. Although this model was developed for forest ecosystems, we also applied it on the heathland data. This approach seems justified since Bobbink et al (1992b) did find a good correlation between the amounts of cation loss from the canopy with the observed ammonium uptake by *Calluna*. The calculations indicate highest canopy exchange values of Ca^{2+} in the case of grassed heathland and of Mg^{2+} in the case of pure heathland, explaining the patterns observed in throughfall deposition.

5.5.2 Do vegetation types differ in their seepage quantities?

The differentiation between the different vegetation types for throughfall deposition was not univocally reflected in the global clustering of ion seepage flux. Heathland was clearly distinguished from forest, but no differentiation was made between ion seepage fluxes of different heathland and forest types, except for the plots of *Pinus nigra*. Although no significant differences were found in throughfall deposition of NH_4^+, NO_3^- and SO_4^{2-} between both coniferous forest types, seepage of NO_3^- and SO_4^{2-} was low in all vegetation types except for the two plots with *Pinus nigra*. This is quite exceptional for the Flemish situation, since most case studies show high seepage quantities of nitrate to groundwater under coniferous and to a lesser degree under deciduous forest (De Schrijver et al. 2000, 2004, Neirynck et al.

2002). The high nitrate and sulphate seepage in the *Pinus nigra* plots is furthermore related to significant losses of so-called base cations and aluminium, reducing the buffering potential of the soil.

5.5.3 Are there signs of nitrogen saturation in the studied vegetation types?

Input-output budgets from European forests have shown that above a threshold of approximately 10 $kg.ha^{-1}.yr^{-1}$ of N in throughfall, many sites appear N saturated and have nitrate-seepage rates above 5 $kg.ha^{-1}.yr^{-1}$ (Dise et al. 1998, Gundersen et al. 2006). For our dataset we can conclude that, despite the exceedance of this threshold value, nitrogen retention in all the studied vegetation types other than *Pinus nigra* is sufficiently high to avoid significant nitrate leakage. Furthermore, Gundersen et al. (2006) did found that for all European forest sites receiving more than 25-30 $kg.ha^{-1}.yr^{-1}$ of N in throughfall, elevated nitrate seepage occurred. An important finding was also that for the whole range of N input, both retention and no retention were found, depending on the nitrogen availability of the system (N status) (Gundersen et al. 1998a) and the type of nitrogen input (Emmett et al. 1998a).

Retention of N deposition in soils mainly occurs in the forest floor (Nadelhoffer et al. 2004). For coniferous forest, the forest floor C to N ratio is found to be a good indicator of the N status (Dise et al. 1998, Gundersen et al. 1998a, Kristensen et al. 2004), while this is not the case for deciduous forest having generally thin forest floors (Kristensen et al. 2004, Augusto 2002). MacDonald et al. (2002) found a distinction in nitrogen retention between sites with forest floor C/N ratios above and below 25. This threshold value of about 25 (range between 24-27, Gundersen et al. 2006) was established by several authors (Dise et al. 1998, Gundersen et al. 1998a, Kristensen et al. 2004, Gundersen et al. 2006), although the variability on the data is very high (Dise et al. 1998). The decrease in C/N-ratio of the forest floor seems to be the onset of mineralization and nitrification reactions by fungi and bacteria (e.g. Lovett and Rueth 1999, Goodale and Aber 2001, Aber et al. 2003). Three of the four studied coniferous plots in the present study have forest floor C/N-ratios below this threshold, which indicates a high N status (Gundersen et al. 2006). Furthermore, Dise et al. (1998) and Kristensen et al. (2004) found for coniferous plots that soil solution nitrate concentrations and seepage quantities can be best predicted by a model with the throughfall N deposition as main factor besides organic layer C/N ratio. The *Pinus nigra* plot (Pn2) with highest nitrate seepage is the one with both the highest inorganic nitrogen throughfall input (28.8 kg $N.ha^{-1}.yr^{-1}$) and lowest

C/N-ratio (22). Although our dataset is far too small to validate the model of e.g. Kristensen (2004), we hypothesize that the *Pinus nigra* plots already evolved to the situation of nitrogen saturation, while the plots of *Pinus sylvestris* are still in an intermediate phase between nitrogen limited and nitrogen saturated conditions.

One deciduous (D2) and one *Pinus nigra* (Pn2) plot were different in their historical land use. Both plots were cultivated and used as field or meadow during the period 1771-1850 and for D2 even till 1940. Forests on former agricultural land can have a higher soil nutrient status than old grown forest (Compton and Boone 2000), and therefore are more vulnerable to disturbance of the nitrogen cycle. However, for sandy soils less persistent impacts of agriculture on soil C and N content and N cycling have been found (Compton et al. 1998). Nevertheless, the relatively low C/N ratios in the upper part of the mineral soil might indicate a time-lag of former agricultural use, and may explain why the Pn2 *Pinus nigra* plot shows double the amount of nitrate seepage compared to plot Pn1.

Also for heathland, N addition experiments have shown a reduction in organic soil C/N accompanied by a simultaneous increase in nitrate seepage (Evans et al. 2006), which confirms model predictions indicating a more rapid 'breakthrough' of nitrate where N deposition is high and soil C pool is small. In contrast to coniferous forest ecosystems, a threshold value for C/N is not available for heathland. We expect that a shift towards nitrogen saturation will take longer for these ecosystems than for the *Pinus sylvestris* stands, since lower nitrogen loads via throughfall deposition enter the soil: heathland received annually on average 17 kg less nitrogen per hectare via throughfall deposition compared to coniferous forest. Also Herrmann et al. (2005) found less than 5 $kg.ha^{-1}.yr^{-1}$ nitrate seepage under heathland, while this value was equalled or exceeded under most forest sites. Significant seepage of nitrate under heathland has, however, been reported for Dutch heathland ecosystems receiving high levels of atmospheric N deposition for several decades (Schmidt et al. 2004) which is, according to Kristensen and Henriksen (1998), attributable to the high net N mineralization rate and occurrence of net nitrification.

5.5.4 Is there a future risk for nitrogen saturation in the studied vegetation types?

It is very difficult to make quantitative predictions of the future risk of increased N seepage from both heathland (Evans et al. 2006) and forest ecosystems (Gundersen et al. 2006). Soil carbon and nitrogen pools and the current and past N deposition load seem to be the

determining factors, but interactions between these are poorly understood for the whole range of soil types (Gundersen et al. 2006). Our dataset implies that the choice of vegetation type may delay the time of onset of nitrogen saturation and its negative consequences. Nowadays, coniferous plantations are conversed towards mixed forests with broadleaves from the perspective of forest management (Spiecker et al. 2004), while nature conservation strives for extension and reconnection of heathland fragments. One might expect that heathland and all types of forest ecosystems receiving high loads of N during several decades will show elevated nitrate seepage to groundwater on the long-term (Schmidt et al. 2004, Gundersen et al. 2006). Comparisons of seepage quantities under heathland and forest ecosystems subjected to high nitrogen load are lacking, so well-considered advice on optimal ecosystem choice in terms of protection of groundwater can not be made till now. Converting coniferous to deciduous forest is, however, expected to cause improvements since at the status of nitrogen saturation, the ecosystem is very responsive to changes in N deposition (Gundersen et al. 1998b). Reductions caused by a change in tree species composition will therefore generally result in a significant decrease in nitrate seepage (De Schrijver et al. in press), with an undefined time delay because of accelerated mineralization and nitrification at the time of conversion. It must, however, be clear that conversion of coniferous forest to any other vegetation type will only delay the onset of nitrogen saturation, as the source of the problem (high nitrogen emissions) is still occurring in large parts of Europe.

5.6 Conclusions

The division of vegetation types in separate clusters shows that each vegetation type has its specific characteristics influencing the throughfall input and seepage to groundwater of pollutants/nutrients. Seepage of pollutants to groundwater is limited for pure and grassed heathland, deciduous forest and stands of *Pinus sylvestris*, while *Pinus nigra* stands show elevated seepage of nitrate, sulphate, so-called base cations and aluminium. The *Pinus nigra* plots clearly evolved to the situation of nitrogen saturation, while the other vegetation types are still in an intermediate phase between nitrogen limited and nitrogen saturated conditions.

6 Forest type effect on throughfall deposition and seepage flux in an area with high deposition load

After: De Schrijver A., Van Hoydonck G., Nachtergale L., De Keersmaeker L., Mussche S. & Lust N. (2000). Comparison of nitrate seepage under silver birch (*Betula pendula*) and Corsican pine (*Pinus nigra ssp. laricio*) in Flanders (Belgium). Water, Air and Soil Pollution, 122, 77-91.

De Schrijver A., Nachtergale L., Staelens J., Luyssaert S. & De Keersmaeker L. (2004). Comparison of throughfall and soil solution chemistry between a high-density Corsican pine stand and a naturally regenerated silver birch stand. Environmental Pollution 131, 93-105.

6.1 Abstract

In Flanders, critical loads for acidification and eutrophication are exceeded in the majority of the forest stands, and many previously nitrogen limited forest ecosystems have become nitrogen saturated. The present study investigates whether a naturally regenerated stand of silver birch (*Betula pendula* Roth) contributes less to the acidification and eutrophication of the forest soil than a high-density plantation of Corsican pine (*Pinus nigra ssp. laricio* Maire) in a region with high emissions from intensive livestock husbandry. Throughfall deposition of inorganic nitrogen was about 3.5 times higher in the Corsican pine stand than in the birch stand. Potassium throughfall deposition was significantly higher under birch due to higher canopy leaching. Magnesium throughfall deposition was significantly higher under the pine canopy due to higher dry deposition.

The lower nitrogen throughfall deposition in the birch stand was reflected in a 60% lower nitrate seepage at 1 m depth compared with pine. Nitrate seepage is linked to losses of aluminium and so-called base cations. Our results can contribute to forest management, since a well-considered tree species-choice in forests can help diminishing soil acidification and nitrate seepage to groundwater.

6.2 Introduction

With a forest cover of less than 10%, the northern part of Belgium (Flanders) has one of the lowest afforestation rates of Europe (Luyssaert et al. 1999). About 30% of the Flemish forest consists of first or second generation Scots pine (*Pinus sylvestris* L.) and Corsican pine (*Pinus nigra ssp. laricio* Maire) stands, located on sandy soils which were formerly covered by heath. Most of these forests are homogeneous plantations on podzol soils with low buffering capacity, and have shown to be susceptible to infestations (like *Sphaeropsis sapinea* (Fr.) Dyko & Sutton).

The condition of these forests has become even more critical due to increasing stress from high atmospheric deposition levels. The high exogenous input of nitrogen and sulphur is due to (*i*) high concentrations of atmospheric pollutants (Van Avermaet et al. 2006), (*ii*) high humidity, which increases the collection efficiency for airborne material (Ruijgrok et al. 1997) and (*iii*) a high degree of forest fragmentation leading to a reinforcement of edge deposition effects (Draaijers 1988). As a result, the deposition of nitrogen and sulphur in Flanders exceeds the critical loads for acidification and eutrophication in the majority of forest stands (Overloop et al. 2006; Van Avermaet et al. 2006).

Forest structure, canopy density and tree species composition were reported to have a significant impact on dry deposition. Dry deposition is increasing with increasing tree height (Lovett and Reiners 1986) and increasing leaf area index (Ivens 1990). Dry deposition is furthermore dependent on tree species composition, as conifers are found to be more efficient in collecting particles and cloud droplets compared to broadleaf trees (Brown and Iles 1991 Draaijers 1988). Also nutrient exchange processes in the canopy (Alcock and Morton 1985), uptake capacity by roots (Cole and Rapp 1981; Magill et al. 1997), and litter decomposition rate (Howard and Howard 1990; Johansson 1995) were reported to depend on forest structure and composition.

This study's hypothesis is that, given the high atmospheric nitrogen and potentially acidifying deposition in a major part of Flanders, naturally regenerated forests of broadleaf trees contribute less to soil acidification and eutrophication than homogeneous plantations of coniferous trees. Forest conversion from these high-density plantations to deciduous stands is therefore expected to have an impact on the forest soil by decreasing nitrate seepage, loss of so-called base cations and mobilisation of toxic aluminium. This hypothesis is tested by comparing biogeochemical fluxes through (*i*) throughfall deposition, (*ii*) litterfall, and (*iii*) soil seepage,

between a high-density plantation of Corsican pine and a naturally regenerated stand of silver birch.

6.3 Materials and methods

6.3.1 Site description

The experimental site is located in the 'Hoogmoerheide' nature reserve in Merksplas (51°15′ N, 4°54′ E), in the north-eastern part of Belgium (Flanders). Hoogmoerheide has a surface area of 105 ha and is mainly dominated by heath, homogeneous plantations of Corsican pine (*Pinus nigra ssp. laricio* Maire), homogeneous plantations of Scots pine (*Pinus sylvestris* L.), and naturally developed stands of silver birch (*Betula pendula* Roth*)*. Neighbouring stands of Corsican pine and silver birch were selected. Both stands had about the same age and were growing on the same parent material. The management history of the stands was also very similar. Following the FAO classification (FAO 1988), the coarse sandy soils were classified as Haplic podzols.

The history of the stands was reconstructed using historical topographic maps. In 1850, the vegetation was transformed from heath to Scots pine. Scots pine was harvested and the stand was replanted with Corsican pine around 1955. Between 1955 and 1985, the birch stand developed naturally as the sublayer of a Scots pine stand. In 1985, the Scots pines were cut and the birches became the dominant tree species.

Both stands are about 40 years old. In 1999, average tree height, basal area and stem volume were 14.4 m, 16 m^2 ha^{-1} and 111 m^3 ha^{-1} for the silver birch stand, and 16.1 m, 45 m^2 ha^{-1} and 365 m^3 ha^{-1} for the Corsican pine stand. These stand characteristics are representative for Flanders and the Netherlands (Jansen et al. 1996). The leaf area index (LAI) of the pine stand was visually estimated to be three times higher than the LAI of the birch stand.

6.3.2 Experimental set-up

The experiment was set up as three replicated plots under Corsican pine and silver birch. To avoid a forest edge effect on the deposition values (Draaijers et al. 1988; De Schrijver et al. 1998), all plots were established at a fixed distance of 50 m from the forest edge. Each plot was equipped with four throughfall collectors, suction lysimeters with ceramic cup at three depths (0.1 m, 0.5 m and 1 m), and five circular litter traps of 0.3 m^2. The applied suction on

the porous cup lysimeters was –50 kPa. Bulk deposition was collected using four bulk collectors placed above the heath adjacent to the forest. Throughfall and bulk precipitation were collected using polyethylene funnels (15 cm diameter) supported by and draining into two-litre polyethylene bottles. The bottles were placed below ground level to avoid the growth of algae and to keep the samples cool. A nylon wire mesh was placed in the funnels to prevent contamination by large particles. Stemflow water was not collected because of its low contribution to nutrient fluxes in young pine and birch stands.

Water fractions were collected and measured fortnightly from September 1998 to April 1999 and monthly from May 1999 to February 2000. On each sampling occasion, the water volume in every collector was measured in the field, and the bottles were replaced by bottles rinsed with distilled water. The four throughfall samples of each plot were pooled to one sample for the chemical analyses. All water samples were transported and stored at a maximum temperature of 5°C. After the samples had been analysed for pH (ion-specific electrode), they were filtered through a glass microfibre filter (WHATMAN GF/A) and a nylon membrane filter (GELMAN, nyloflo) of 0.45 μm. Samples were analysed within a week for Cl^- (ion-specific electrode), NO_3^- (UV photometric method at λ=210 nm according to the Dutch standard method NEN 6581), NH_4^+ (photometric determination of a reaction product of NH_4^+ at λ=660 nm according to the Dutch standard method NEN 6576), and K^+, Ca^{2+}, Mg^{2+}, Na^+ and Al(III) (flame atomic absorption spectrophotometry).

Litterfall was collected fortnightly between September 1998 and January 1999, and dry litterfall weight was determined after drying for 48 h at 70°C. Litterfall samples were analysed on total N (modified method of Kjeldahl) and K^+, Ca^{2+}, Mg^{2+} and Na^+ (flame atomic absorption spectrophotometry) after oxidation with a HNO_3 digestion.

Mineral soil samples were taken on different depths (0-5, 5-15, 15-30, 30-60 and 60-100 cm), dried, sieved and analysed on pH (total N (modified method of Kjeldahl), C (Walkey & Black) and exchangeable amount of cations K^+, Ca^{2+}, Mg^{2+}, Na^+ and Al (III) using flame atomic absorption spectrophotometry after extraction with NH_4-EDTA (Table 6.1). Samples from the forest floor were taken and analysed on total C (method of Springer and Klee 1953) and total N (Kjeldahl digestion) by Vervaet et al. (2003).

Table 6.1: Some mineral soil characteristics of the silver birch and Corsican pine stands at Merksplas, 1999

Soil depth *(cm)*	**pH-H_2O**		**Exchangeable Mg^{2+}** *(meq/100 g)*		**Exchangeable Ca^{2+}** *(meq/100 g)*		**Exchangeable Al (III)** *(meq/100 g)*	
	Birch	Pine	Birch	Pine	Birch	Pine	Birch	Pine
0 – 5	3.7	3.6	0.27	0.18	1.07	0.75	6.63	12.15
5-15	3.9	3.7	0.09	0.07	0.48	0.27	7.25	23.11
15-30	3.8	3.8	0.05	0.04	0.29	0.13	25.16	35.67
30-60	3.9	4.1	0.03	0.01	0.15	0.04	44.25	26.26
60-100	4.1	4.3	0.02	0.01	0.08	0.02	48.35	17.53

6.3.3 Quality control

The quality of the chemical analyses was checked by including method blanks, repeated measurements of internal and certified reference samples, and by inter-laboratory tests. The methods were validated with samples from inter-laboratory profession tests and standard reference materials (CRM 100, CRM 409). For water samples the relative standard deviation (Stdev/average*100) based on repeated measurements of internal quality controls was 4.6% for NH_4^+, 3.3% for NO_3^-, 5.1% for Na^+, 4.3% for K^+, 4.1% for Ca^{2+}, 4.3% for Mg^{2+}, and 4.7% for Al(III). For plant samples the relative standard deviation based on repeated measurements of CRM 100 was 0.8% for N, 4.4% for K, 3.5% for Ca and 4.6% for Mg.

6.3.4 Calculation of throughfall deposition and litterfall fluxes

Ion bulk and throughfall deposition was calculated by multiplying the water volume with the element concentration in that volume. Nutrient input by litterfall was calculated by multiplying the amount of litterfall with the nutrient concentration in the litter.

6.3.5 Calculation of canopy exchange quantities

Compared to bulk precipitation, the chemical composition of throughfall water ions is generally altered, and it is widely acknowledged that this transformation results from (*i*) the washing off of dry deposition of aerosols and gases, as well as (*ii*) canopy leaching, i.e. release of ions from plant tissues or (iii) canopy uptake (Parker 1983). The calculation method is described in §5.3.4.

Additionally, we estimated canopy uptake of ammonium and protons by assuming that the leaching of so-called base cations from the canopy equals the uptake of protons and ammonium in a proportion equal to their fluxes in bulk deposition and throughfall (De Vries et al. 1998). The canopy uptake (CU) of H^+ and NH_4^+ is subsequently calculated from the sum of the exchanged cations of K^+, Ca^{2+} and Mg^{2+} (Equation 6.1)

$$CU_{NH_4} = \frac{(BD+TF)_{NH_4}}{(BD+TF)_{NH_4}+(BD+TF)_{H}} \cdot CL_{K+Ca+Mg} \quad (6.1)$$

Knowing the canopy uptake of NH_4^+, the dry deposition flux of NH_4^+ (NH_3 and NH_4^+ aerosol) can be computed from $TF + CU - BD$.

Although uptake of NO_x in the crown is reported for both coniferous and deciduous forest types (Mahendrappa 1989, Neary and Gizyn 1994, Harrison et al. 2000), calculations were not performed in the absence of a calculation method based on throughfall measurements. Canopy uptake was therefore assumed to be low (Neary and Gizyn 1994), allowing a rough calculation of dry deposition of NO_3^- (NTW-bulk deposition (see also §5.3.4).

6.3.6 Calculation of seepage flux

Ion seepage fluxes were calculated by multiplying the calculated water percolation volume at 1 m depth with the average ion concentration in the soil solution at 1m depth. Since meteorological parameters were not measured in the stands, the percolation water flux at 1 m depth was calculated using the Chloride Mass Balance (CMB) method, but with Na^+ instead of Cl^- as a tracer. The calculation method is described in §5.3.4.

6.3.7 Statistical analysis

As water fractions were collected and measured fortnightly from September 1998 to April 1999, but monthly from May 1999 to February 2000, the fortnightly deposition data were summed and the fortnightly data of soil solution concentrations were averaged on a volume-weighted basis.

To compare the ion fluxes of both tree species throughout time, a repeated measures test (also called: a within-subjects test) was performed (for more explanation see §5.3.4). Statistical analyses were performed on monthly data for the growing season (May to November; n = 10)

and the dormant season (December to April; n = 8), as well as for the complete measuring period (n = 18). To allow comparison between nutrient inputs via throughfall deposition and litterfall, the annual data of September 1998 to August 1999 are presented.

6.4 Results

6.4.1 Hydrologic fluxes

During the 18-month measuring period, total bulk precipitation was 1538 mm. Average interception by the canopy represented 17% of the total precipitation for the deciduous stand and 37% for the coniferous stand. This resulted in a significantly different ($p < 0.001$) total throughfall amount of 1250 mm in the silver birch stand and 961 mm in the Corsican pine stand.

Using Cl^- as a tracer (data not shown), the calculated quantity of percolation water under both stands was comparable. Percolation output was somewhat higher under the birch stand (541 mm) than under the pine stand (489 mm), which corresponds to the throughfall quantities.

6.4.2 Throughfall deposition and litterfall fluxes

The bulk and throughfall deposition of nitrogen consisted mainly of ammonium (Fig. 6.1, Table 6.2 and Table 6.3). Throughfall nitrogen deposition was 1.5 times higher than bulk deposition in the birch stand, and five times higher than bulk deposition in the pine stand (Fig. 6.1 and Table 6.3). Throughfall ammonium and nitrate deposition in the pine stand were significantly higher than in the birch stand (Table 6.2). Moreover, nitrogen deposition was significantly influenced by time, indicating a strong seasonality in N deposition (Fig. 6.1). For the total measuring period, a significant interaction was found between tree species and time concerning N deposition.

In both stands, throughfall deposition of ammonium-N mainly resulted from dry deposition (Table 6.3). According to the canopy budget model, 471 $mol.ha^{-1}.yr^{-1}$ and 150 $mol.ha^{-1}.yr^{-1}$ ammonium-N were retained in the canopies of the birch and the pine stands respectively (Table 6.3), especially during the growing season (not shown). Nitrate-N mainly entered the birch stand by bulk deposition, and only about 30% originated from dry deposition. In the pine stand on the other hand, throughfall deposition of nitrate-N was mainly the result from dry deposition, and only 29% originated from bulk deposition.

Input of N via litterfall was 1.8 kmol.ha^{-1}.yr^{-1} in the birch stand and 2.3 kmol.ha^{-1}.yr^{-1} in the pine stand (Table 6.3). Adding inorganic nitrogen throughfall deposition and total nitrogen input via litterfall gave 3.8 kmol.ha^{-1}.yr^{-1} in the birch stand and 9.2 kmol.ha^{-1}.yr^{-1} in the pine stand.

Table 6.2: Average monthly bulk and throughfall depositions (± standard deviation) expressed in mol.ha^{-1}, during the growing season, the dormant season and the whole year for the measuring period (September 1998 - February 2000). Significance levels of species (Sp.), time (Ti.) and interaction between species and time (Int.) are indicated (ANOVA, repeated measures design)

	Growing season				**Dormant season**				**Study period**			
	Mean ± sd.	Sign.			Mean ± sd.	Sign.			Mean ± sd.	Sign.		
		Sp.	Ti.	Int.		Sp.	Ti.	Int.		Sp.	Ti.	Int.
Ammonium-N												
Bulk	108 ± 69				73 ± 54				93 ± 64			
Silver birch	135 ± 90				113 ± 70				126 ± 81			
Corsican pine	423 ± 271	*	***	***	296 ± 184	**	***	**	369 ± 241	**	***	***
Nitrate-N												
Bulk	27 ± 17				13 ± 9				21 ±15			
Silver birch	40 ± 18				22 ± 14				32 ± 18			
Corsican pine	80 ± 34	**	***	***	50 ± 34	**	***	*	67 ± 37	**	***	***
Potassium												
Bulk	11 ± 14				4 ± 3				8 ± 11			
Silver birch	53 ± 41				14 ± 12				37 ± 38			
Corsican pine	36 ± 28	**	***	**	22 ± 16	ns.	***	***	30 ± 24	*	***	***
Calcium												
Bulk	15 ± 7				18 ± 14				16 ± 11			
Silver birch	20 ± 11				18 ± 15				19 ± 12			
Corsican pine	21 ± 15	ns.	***	**	24 ± 20	ns.	***	***	22 ± 17	ns.	***	***
Magnesium												
Bulk	7 ± 5				9 ± 12				8 ± 8			
Silver birch	12 ± 9				12± 12				12 ± 10			
Corsican pine	14 ± 12	ns.	***	ns.	16 ±12	*	***	***	15 ± 12	*	***	**
Sodium												
Bulk	112 ± 103				108 ± 89				110 ± 95			
Silver birch	132 ± 105				133 ± 103				132 ± 101			
Corsican pine	160 ± 155	ns.	**	ns.	170 ± 117	ns.	***	ns.	169 ± 136	ns.	***	ns.
Protons												
Bulk	4 ± 4				7± 6				5 ±5			
Silver birch	2 ± 2				5±5				4 ±4			
Corsican pine	1 ± 1	**	***	***	1± 1	***	***	**	0 ±1	***	***	***

* P < 0.05, ** P < 0.01; *** P < 0.001

Throughfall deposition of Ca^{2+}, Mg^{2+} and K^{+} was higher than bulk deposition during both growing and dormant seasons (Fig. 6.1, Table 6.2). Relative to bulk deposition and taken over

a period of a whole year, the Ca^{2+}, Mg^{2+} and K^{+} throughfall depositions were enriched by a factor 1.2, 1.6 and 5 respectively under the deciduous canopy, and by 1.4, 1.9 and 3.4 respectively under the coniferous canopy. Throughfall deposition of K^{+} and Mg^{2+} was significantly different between the birch and pine stand for the whole measuring period (Table 6.2). A significant tree species effect was also observed for K^{+} during the growing season and for Mg^{2+} during the dormant season. Throughfall deposition of all so-called base cations was significantly affected by time (Fig. 6.1). The interaction between tree species and time was significant for Ca^{2+}, Mg^{2+} and K^{+}.

Throughfall deposition of protons was generally lower than bulk deposition. The repeated measures design revealed a significant effect of tree species and time on throughfall deposition of protons (Table 6.2) for the whole year. Both forest canopies neutralized significant amounts of protons, especially during the growing season. Birch and pine retained 34% and 82% of the incoming protons, respectively.

Table 6.3: Bulk deposition, throughfall deposition split up calculated dry deposition and canopy leaching (+) / uptake (-), input of nutrients via litterfall, and total input to the forest floor (throughfall deposition + litterfall) for the period September 1998-August 1999. All fluxes in $mol.ha^{-}1.yr^{-1}$

	Bulk deposition	**Dry deposition**	**Leaching / uptake**	**Throughfall deposition**	**Litterfall**	**Total input**
Birch stand						
Ammonium-N	900	1114	-471	1543	-	-
Nitrate-N	314	136	/	450	-	-
Total nitrogen	1214	1250	-471	1993	1779	3772*
Potassium	118	31	445	594	128	722
Calcium	247	67	-7	307	140	447
Magnesium	107	29	37	173	91	264
Pine stand						
Ammonium-N	900	5057	-150	5807	-	-
Nitrate-N	314	771	/	1085	-	-
Total nitrogen	1214	5828	-150	6892	2279	9171*
Potassium	118	74	212	404	95	499
Calcium	247	152	-57	342	157	499
Magnesium	107	66	29	202	49	251

* Throughfall deposition of inorganic nitrogen + total nitrogen in litterfall

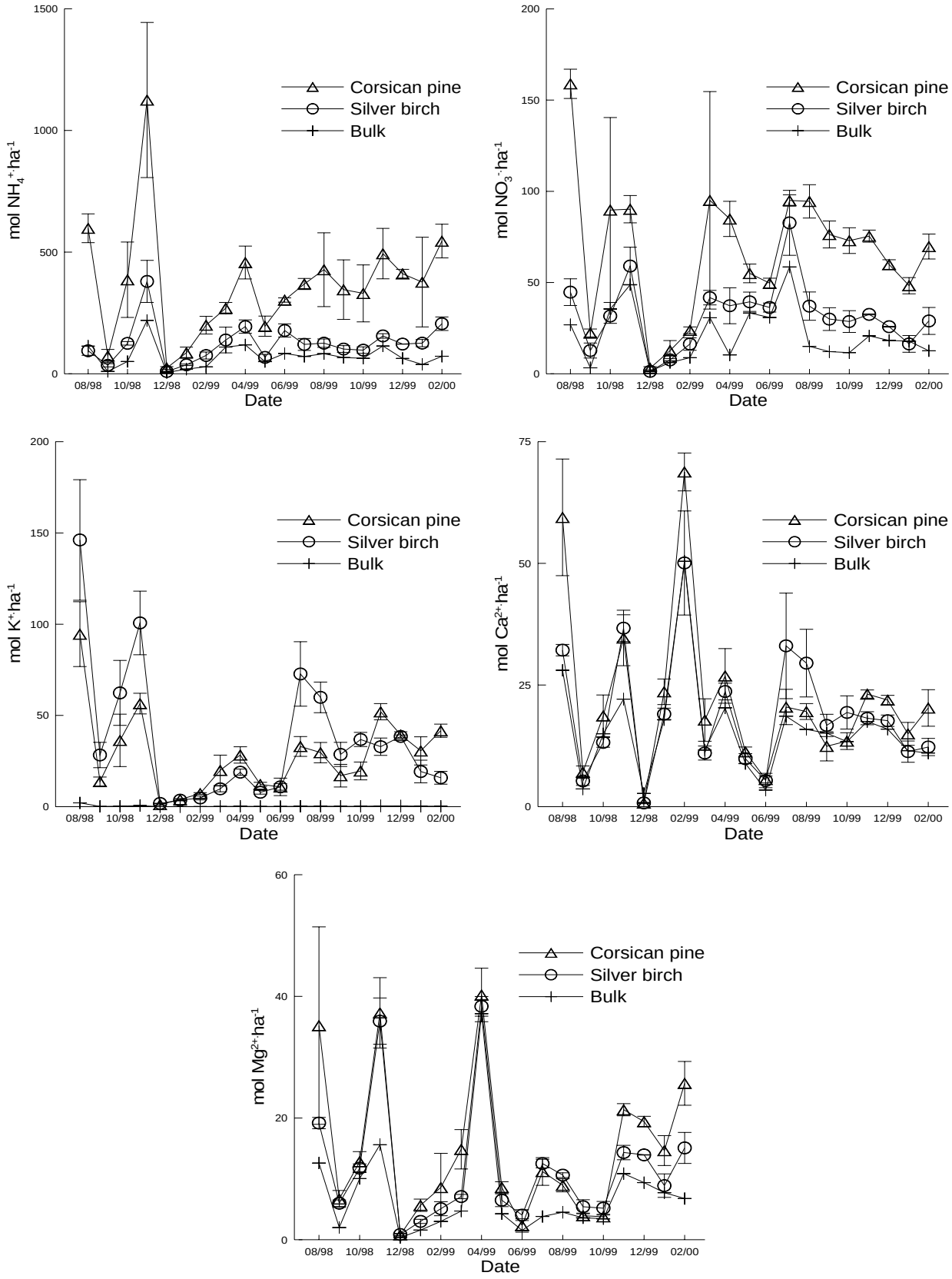

Fig. 6.1: Bulk and throughfall deposition (mol.ha^{-1}.month^{-1}) of NH_4^+, NO_3^- K^+, Ca^{2+}, and Mg^{2+} in the Corsican pine plantation and the silver birch stand.

6.4.3 Seepage flux

In contrast to throughfall water, seepage fluxes at 1 m depth were dominated by nitrate and not by ammonium (Table 6.4). Nitrate concentrations in the soil solution at 1 m depth were significantly influenced by both tree species and time (Fig. 6.2, Table 6.5) for the whole year as well as for the dormant and the growing season separately. Compared to the soil solution under birch, nitrate concentrations under pine were on average three times higher in the growing season and two times higher in the non-growing season. Only time was of significant influence on the low NH_4^+ soil solution concentrations.

Concentrations of K^+, Ca^{2+}, Mg^{2+} and Al(III) in the soil solution at 1 m depth were clearly time and species dependent (Table 6.5). Tree species significantly influenced the soil solution concentration of so-called base cations and aluminium, annually and on a seasonal basis, except for K^+. On a yearly basis, the interaction between tree species and time was significant for the base cations, but not for Al(III). For the dormant season, K^+, Ca^{2+} and Mg^{2+} concentrations in the soil solution were significantly affected by tree species, time and the interaction between these two factors. Total seepage flux of all so-called base cations and aluminium was higher under Corsican pine than under silver birch (Table 6.4). Proton concentrations in the soil solution at 1 m depth were only time dependent. No significant differences between species were found.

Table 6.4: Ion seepage (mol.ha^{-1}.period^{-1}) at 1 m depth under the Corsican pine and the silver birch stand

	September 1998-August 1999		**September 1999-February 2000**	
	Birch	Pine	Birch	Pine
Ammonium-N	107	107	0	14
Nitrate-N	1814	4029	443	1600
Potassium	141	159	59	59
Calcium	517	1187	130	431
Magnesium	173	239	21	66
Aluminium	878	1907	341	678
Protons	200	200	100	100

Table 6.5: Average monthly concentrations (µmol.l^{-1}) and standard deviation of ions in the soil solution at 1 m depth during growing season, dormant season and the whole year for the measuring period (September 1998 - February 2000). Significance levels of species (Sp.), time (Ti.), and interaction between species and time (Int.) are indicated (ANOVA, repeated measures design)

	Growing season				**Dormant season**				**Study period**			
	Mean ± sd.	Sign.			Mean ± sd.	Sign.			Mean ± sd.	Sign.		
		Sp.	Ti.	Int.		Sp.	Ti.	Int.		Sp.	Ti.	Int.
Ammonium-N												
Silver birch	7 ± 7	ns.	*	ns.	19 ± 26	ns.	**	ns.	15 ± 23	ns.	***	ns.
Corsican pine	5 ± 36				18 ± 30				13 ± 26			
Nitrate-N												
Silver birch	453 ± 258	***	***	***	460 ± 165	**	**	ns.	456 ± 216	***	***	***
Corsican pine	1466 ± 558				800 ± 144				1150 ± 523			
Potassium												
Silver birch	46 ± 20	ns.	ns.	ns.	31 ± 23	ns.	***	***	38 ± 22	*	***	***
Corsican pine	59 ± 31				41 ± 33				49 ± 33			
Calcium												
Silver birch	117 ± 90	***	***	***	45 ± 33	*	***	ns.	132 ± 80	**	***	***
Corsican pine	294 ± 347				66 ± 37				299 ± 267			
Magnesium												
Silver birch	33 ± 29	**	***	***	45 ± 33	**	*	**	39 ± 30	**	***	***
Corsican pine	58 ± 41				66 ± 37				62 ± 41			
Aluminium												
Silver birch	296 ± 107	**	***	*	229 ± 78	ns.	ns.	ns.	263 ± 96	*	***	ns.
Corsican pine	607 ± 278				430 ± 207				515 ± 256			
Protons												
Silver birch	41 ± 13	ns.	***	ns.	84 ± 21	ns.	***	ns.	60 ± 17	ns.	***	ns.
Corsican pine	32 ± 13				69 ± 26				50 ± 20			

* P < 0.05, ** P < 0.01; *** P < 0.001

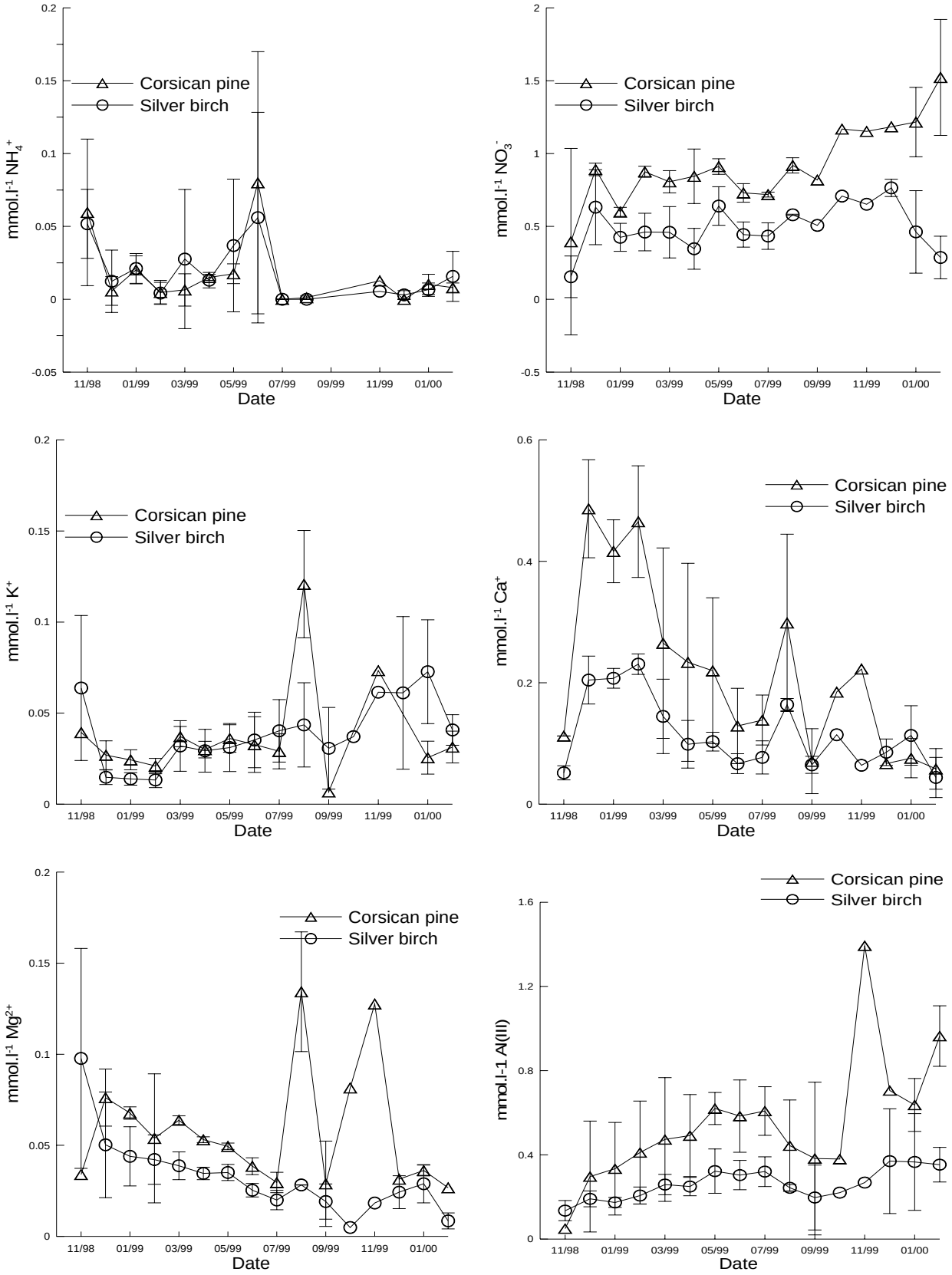

Fig. 6.2: Concentrations (in mmol.l^{-1}) of NO_3^-, NH_4^+, K^+, Ca^{2+}, Mg^{2+} and Al(III) in the soil solution at 1 m depth

6.5 Discussion

Compared to bulk deposition, a net NH_4^+ and NO_3^- enrichment in throughfall water was observed for both stands. In the birch stand, throughfall deposition of inorganic nitrogen was 3.5 times lower than in the pine stand. Von Wilpert et al. (2000) found that the deposition load in deciduous beech stands was 45-85 % lower than in spruce stands. In the Netherlands, nitrogen deposition was significantly higher in stands of Douglas fir and Scots pine than in oak stands (Van Ek and Draaijers 1994). Differences in stand and crown characteristics, such as lower stand and crown density, lower LAI, and the summergreen character, are important factors for lower throughfall ion fluxes in deciduous forest stands (Alcock and Morton 1985, Draaijers 1993, Houle et al. 1999, Augusto et al. 2002).

6.5.1 Are ions released from or retained in the canopy?

Nitrogen

According to the canopy budget method, ammonium was retained by the canopy of both forest types, but to a greater extent in the birch stand than in the pine stand. The performed calculations are based on the assumption that total canopy uptake of protons and ammonium is equal to the total canopy leaching of Ca^{2+}, Mg^{2+} and K^+ due to cation exchange. The reliability of the calculated ammonium uptake by the canopy depends on (i) the calculated so-called base cation leaching, and (ii) the relationship between canopy leaching of base cations and canopy uptake of ammonium and protons. The reliability of the estimated base cation leaching will be discussed later. The calculated ammonium uptake might be overestimated since no corrections were made for passive cation leaching, which may be due to senescence and sprouting or other possible metabolic processes. However, the estimated canopy uptake is in line with literature data: the aboveground uptake of inorganic nitrogen by forests was suggested to be between 150 and 350 $mol.ha^{-1}\ yr^{-1}$ (Ivens 1990), and between 200 and 300 up to 850 $mol.ha^{-1}.yr^{-1}$ (Lovett 1992). It is not yet clear if higher deposition results in a higher uptake rate (Bleeker and Draaijers 2002). Although nitrogen saturation in the canopy might be expected, the canopy uptake of nitrogen is governed by many other factors such as the tree's nitrogen demand and the soil N pool. Our results contrast with the research of Neary and Gizyn (1994) and Houle et al. (1999) who found NH_4^+ retention to be higher at a coniferous site than at a deciduous site, in regions with a much lower atmospheric NH_4^+ deposition. Brumme et al. (1992) and Harrison et al. (1991), however, reported faster uptake rates for

nitrogen in the canopy of beech and birch compared to spruce. According to Harrison et al. (2000), these differences may be attributable to a combination of thinner cuticles and higher wettability of foliage and bark of beech and birch.

Protons

The net throughfall flux of H^+ was negative for both species throughout the measurement period, which indicates a net uptake of protons in the canopy and demonstrates the ability of both forest canopies to neutralize potentially acidifying input. Since both forest types receive similar wet-only atmospheric loads and are subjected to similar ecological and meteorological conditions, tree species must be the main regulating factor of neutralization (Mahendrappa 1989). Dry deposition of atmospheric ammonia leads to the consumption of protons, and is much lower for the birch canopy than the pine canopy. This partly explains why proton deposition to the forest floor is significantly higher in the birch stand. The lower acid neutralizing capacity of birch compared to pine could also be due to the canopy exchange of NH_4^+ for H^+ (Neary and Gizyn 1994), as birch retains more NH_4^+ in the canopy.

So-called base cations

For both forest types, canopies were sources of so-called base cations during the growing and dormant seasons. This can be attributed to the washing off of dry deposition (e.g. soil dust) captured by the canopy, and to ions leaching from the leaves. Factors that possibly regulate canopy leaching of so-called base cations are (*i*) precipitation characteristics such as duration (Lovett and Lindberg 1984), incidence (Lovett and Schaefer 1992) and acidity of rain events (Schaefer et al. 1988, Schaefer and Reiners 1990, Hansen et al. 1994), (*ii*) canopy characteristics such as above-ground biomass, nutrient amount and concentrations in tree leaves (Lovett et al. 1989) and (*iii*) forest soil characteristics such as extractable amount of so-called base cations and soil solution characteristics (Lovett and Schaefer 1992).

According to the canopy budget calculation model, the foliar leaching of potassium accounted for 75% and 53% of throughfall deposition in the silver birch and the Corsican pine stands respectively. Houle et al. (1999), Van Ek and Draaijers (1994), Parker (1983) and Ragsdale et al. (1992) also reported that the leaching of K^+ was very important, and generally over 70% of throughfall deposition. Potassium throughfall deposition was significantly higher under birch, which was mainly due to higher leaching during the growing season. Houle et al. (1999), Van

Ek and Draaijers (1994) and Alcock and Morton (1985) also found that deciduous canopies leached significantly more K^+ than coniferous canopies. These differences were most explicit during the growing season, which can be attributed to the higher physiological activity of both forest types in this season.

No significant difference in calcium throughfall deposition between both forest types was found in our case study. According to the canopy budget model, Ca^{2+} was taken up by both canopies, and mostly by the pine canopy. It is probable that the calculated low uptake rates of Ca^{2+} should be seen as bias on the filtering approach, since in literature only leaching is reported (Houle et al. 1999; Lovett and Schaefer 1992). The major assumptions and weakness of the canopy budget model of Ulrich (1983d) can be summarized as (i) the use of bulk deposition instead of wet-only deposition, (ii) the assumption that Mg^{2+}, Ca^{2+} and K^+ containing particles are deposited with equal efficiency as particles containing Na and (iii) the assumed relation between wet and dry deposition of particles. In spite of these assumptions, Draaijers et al. (1998) stated that canopy exchange and dry deposition of Mg^{2+}, Ca^{2+} and K^+ can be estimated reasonably well using throughfall and precipitation measurements in association with the canopy budget model.

Magnesium throughfall deposition was significantly higher under the pine canopy due to the higher calculated dry deposition. The calculated contribution of canopy leaching to total throughfall deposition of Mg^{2+} (21% for birch and 14% for pine) was between that of K^+ and Ca^{2+}, and therefore agrees with previous research (Houle et al. 1999; Van Ek and Draaijers 1994; Ragsdale et al. 1992).

The leaching of K^+ and Mg^{2+} and the uptake of NH_4^+ were highest for the birch canopy, despite its summergreen character and the lower basal area and stem volume of the birch stand. As the meteorological and soil conditions were similar, the higher canopy leaching for birch might be due to the thinner cuticles and higher wettability and the higher nutrient concentrations in the birch leaves compared to pine needles (see later Table 6.7, Johansson 1995). The concentration gradient between water on the surface and the interior of the leaves modifies the degree of uptake or leaching (Schaefer et al. 1988; Lovett et al. 1989; Schaefer and Reiners 1990).

6.5.2 Comparison of input through throughfall deposition and litterfall

In both forest types, nitrogen input through litterfall was lower than the deposition load. In

forests less polluted by atmospheric depositions, input by litterfall is by far the dominant nitrogen source (Ukonmaanaho and Starr 2002). This emphasizes the high degree of disturbance by external pollution sources in the forests of the present study. Total annual N input to the forest floor (throughfall deposition and litterfall) was more than 50% lower in the birch stand than in the pine stand. These differences were mainly due to differences in throughfall deposition.

6.5.3 In which ecosystem compartment is this supplied nitrogen retained?

According to literature, soils typically assimilate from three to more than ten times more N deposition than trees do (Nadelhoffer et al 1995, 2004, Tietema et al. 1998, Schleppi et al. 1999). So only small fractions of N inputs, generally much less than 10% of deposition, are likely to be assimilated into woody tissue (C/N-ratio >200) in temperate forests. Also Johnson (1992b) reported that on average 28% of added N was recovered in vegetation compared to 39% in soil. Similarly, Fenn et al. (1998) reviewed 19 studies in the US and Europe and found that 6-33% of labelled ammonium ($^{15}N\text{-}NH_4^+$) and 4–37% of labelled nitrate ($^{15}N\text{-}NO_3$) was recovered in vegetation compared to 30-87% of $^{15}N\text{-}NH_4^+$ and 19-86% of $^{15}N\text{-}NO_3^-$ recovered in soils. These numbers are however only giving insight in the short-term retention of $^{15}N\text{-}NH_4^+$ and $^{15}N\text{-}NO_3^-$ in the woody biomass and soil. Based on a total analysis of the different pools of nitrogen in a Corsican pine stand in Flanders, Neirynck et al. (1998) also found that only 12% of the ecosystem nitrogen pool was stored in its standing crop.

We did an attempt to estimate the nitrogen stored in the aboveground biomass of the Corsican pine and the silver birch stands (Table 6.6) based on the calculated stem volume, average literature values of wood density and wood N concentration (Staelens et al. 2006). For the N content of fresh needles, we adopted the data of Neirynck et al. (1198) from a somewhat older Corsican pine stand on quite comparable soil. The stem volume in the birch stand is much lower than in the Corsican pine stand, but wood nitrogen concentrations are higher, so the nitrogen stock in the wood accounts to 183 kg $N.ha^{-1}$ in the silver birch stand and 224 kg $N.ha^{-1}$ in the Corsican pine stand. For Corsican pine, we estimated the fresh needle nitrogen content to amount 106 kg.ha-1. These calculations are certainly an underestimation of the total N stock in biomass since branches and roots, which are all nutrient-rich fractions, are not taken into account. Nevertheless, it gives an idea of the N stock in the wood and fresh needles compared to the stock in the soil (forest floor and mineral soil, see also Fig. 6.3) compartment, which is obviously much higher (Table 6.6).

Table 6.6: Calculation of nitrogen stored in the stems and fresh needles (for Corsican pine), forest floor and mineral soil (0-100 cm) of both forest types

	Corsican pine	**Silver birch**
Fresh needles (kg.ha^{-1})	106	/
Stems		
Volume (m³/ha)	365	111
Density (kg/m³)	410	660
Mass (kg/ha)	149650	73260
N concentrations (g/kg)	1.5	2.5
N content (kg/ha)	224	183
Forest floor (kg.ha^{-1})	1069	1235
Mineral soil (kg.ha^{-1})	3923	5217

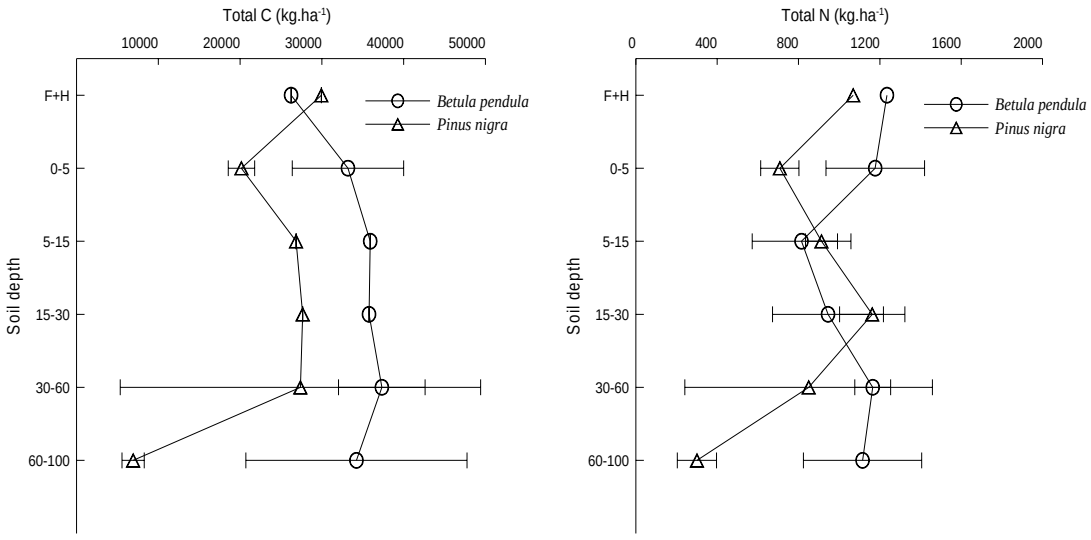

Fig. 6.3: Carbon and nitrogen content (kg.ha^{-1}) in the forest floor and different layers of the mineral soil in the Silver birch and Corsican pine stands

6.5.4 Why do the carbon and nitrogen content of the forest floor and mineral soil layer differ between both species?

Comparing the soils of both forest types, we did find that much more nitrogen and carbon is stored in the soil profile under the silver birch compared to the Corsican pine stand (Fig. 6.3). Contrary to the mineral soil, more C is stored in the forest floor of the Corsican pine stand. Net mineralization of the litter layer, as reported by Vervaet (2003), was found to be

significantly higher in the forest floor of the silver birch stand (13.4 mg $N.m^{-2}.d^{-1}$ compared to 4.2 mg $N.m^{-2}.d^{-1}$in the stand of Corsican pine). A higher net mineralisation rate makes that less carbon and nutrients are stored in the forest floor, higher rates of soil respiration occur (higher CO_2 flux) and more carbon and nutrients can flow through to the underlying mineral layers.

The higher net mineralization rates in the forest floor of the silver birch stand might be related to the quality of its litter (Table 6.7) and furthermore to the lower acidity, higher base saturation and lower aluminium saturation of the soil (Table 6.1) (Fischer and Binkley 2000). Also higher mean annual soil temperatures (Hobbie et al. 2006, Besson et al. 2006) due to a better light penetration to the forest floor (lower LAI) (Hobbie et al. 2006) are influencing mineralization rates. Decomposition of plant material is largely mediated by fungi and bacteria, which have lower C/N ratios than the litter. Litter with high concentrations of N decomposes faster because of the associated faster growth of microbial populations (Cotrufo et al. 2000). In the first stages of litter decomposition, initial litter N content was found to be an important regulating factor, while at later stages lignin concentration becomes a better predictor (Berg et al. 1982). In more recent papers of Reich et al. (2005) and Hobbie et al. (2006), litter calcium, earthworm communities and soil properties of 14 tree species were studied, among which *Pinus nigra* and *Betula pendula*, on a somewhat richer soil type (loamy sand). Trees were planted in blocks after clear cutting a pine plantation. The paper shows a significant negative relationship between litter calcium concentration and earthworm biomass on the one hand and carbon content in the forest floor on the other hand. Silver birch had much more calcium in its litter, had a higher forest floor mineralization rate, a higher soil CO_2 flux and higher earthworm biomass compared to the stand of Corsican pine. These findings are partly in accordance to our results: the silver birch stand had more calcium in its litter (Table 6.7), had a higher forest floor mineralization rate (Vervaet 2003) and less carbon in its forest floor (Fig. 6.3) compared to the Corsican pine stand.

Table 6.7: Characterization of leaf and needle litter in both forest types (Data of lignin (L), hemicellulose (HC) and cellulose (C) are adopted from Vervaet (2003)).

	N	**K**	**Ca**	**Mg**	**L**	**HC**	**C**	**L/N**
	%	*%*	*%*	*%*	*%*	*%*	*%*	
Silver birch	1.92	0.39	0.43	0.17	35.8	10.9	18.9	16.8
Corsican pine	1.23	0.14	0.24	0.05	34.3	13.1	18.1	26

6.5.5 What are the possible mechanisms for nitrogen retention in the soil?

Rapid microbial immobilization has been hypothesized to be the primary mechanism involved in the retention of added dissolved inorganic nitrogen (e.g. Davidson et al. 1992, Hart et al. 1994). However, Tietema (1998) as well as Aber et al. (1998) did not find evidence for gross nitrate immobilization in several forest soils with differing nitrogen availability.

A second possible retention mechanism comprises the abiotic incorporation of ammonium into soil organic matter (Aber et al. 1998) and of nitrate through the so-called 'ferrous wheel' mechanism (Davidson et al. 2003). This hypothesized mechanism implies that abiotic nitrate immobilization occurs through reduction of nitrate to nitrite that subsequently reacts with dissolved organic matter (DOM) to produce dissolved organic N (DON). This DON might be seeped from the ecosystem, retained in soils or converted to plant available forms (Davidson et al. 2003). Estimates of this abiotic nitrogen incorporation indicate that the magnitude of this nitrogen sink is far too small to account for the observed magnitude of nitrogen retention e.g. described for several experimental sites by Aber et al. (1998).

The third proposed mechanism is the assimilation and exudation of nitrogen by mycorrhizal fungi. It was hypothesized by Aber et al. (1998) that mycorrhizae assimilate mineral nitrogen, convert it into extracellular enzymes and exude these enzymes back into the soil, where they could be stabilized in soil organic matter. The mycorrhizae are longer lived than free-living microbes, act as an extension of root systems and are generally thought of as a mechanism by which plants increase the efficiency of nutrient uptake (Treseder et al. 2004). The biomass of mycorrhizal fungi can be especially large under nitrogen-limiting conditions, and may decline with nitrogen saturation (Tietema 1998). Aber (1998) hypothesized that the loss of mycorrhizal function due to the huge availability of nitrogen in the soil can be the key process leading to increased nitrification and nitrate mobility. No recent proofs to the contrary are available, although a recent meta-analysis of Treseder (2004) only found an average (but highly variable) reduction of mycorrhizal abundance by 15% after N fertilization. As was also mentioned in §1.2.1, more research is needed to fully understand the nature, kinetics and capacity of different N retention mechanisms.

6.5.6 How much of the nitrogen throughfall deposition is retained in the two forested ecosystems?

The seepage of nitrate at 1 m depth was high in both forest types, but 60% lower in the birch compared to the pine stand. The nitrate found below the rooting zone may originate from deposited NO_3^- transported through the soil profile or from NO_3^- formed in the soil through mineralization and nitrification reactions. Seepage of ammonium was negligible at this depth in both forest types.

The nitrate seepage in the Corsican pine stand represents 60% of inorganic nitrogen throughfall deposition, while in the silver birch stand a ratio of 1 is approached (Fig. 6.4). Assuming a steady state in the litter layer - so the amount of nitrogen coming in through litterfall approximately equals the amount of nitrogen being mineralized/nitrified from the forest floor on a yearly basis - the birch stand does not retain any nitrogen in the soil. Only the amount taken up by trees for storage in leaves, branches and wood is retained. We calculated a net-ecosystem-retention of 40.7 kg N (of which 2.1 kg uptake in the crown) in the Corsican pine stand and 7.6 kg N (of which 6.6 kg N uptake in the crown) in the silver birch stand (Fig. 6.4).

In reality, part of the nitrogen stored in leaves is translocated in the woody biomass before leave/needlefall. Furthermore, N taken up by the crown will partly be supplied to the forest floor by litterfall, and partly be translocated in the above- or belowground biomass (Harrison et al. 2000).

It is likely, however, that the stand of Corsican pine is not in a steady state phase, but still accumulating litter and thus also nutrients and carbon in its forest floor. This might implicate that on a yearly basis not all nitrogen and other nutrients required for plant uptake are released from the litter layer. To fulfil the plant requirements, additional nitrogen must be taken up originating from atmospheric deposition. It might be possible that the birch plot, because of higher nutrient turnover in its litter layer, more bends to this ‘steady state phase’. The amount of nitrogen coming from the birch leaves might even be complemented by nutrient release from the former build-up forest floor under *Pinus sylvestris*.

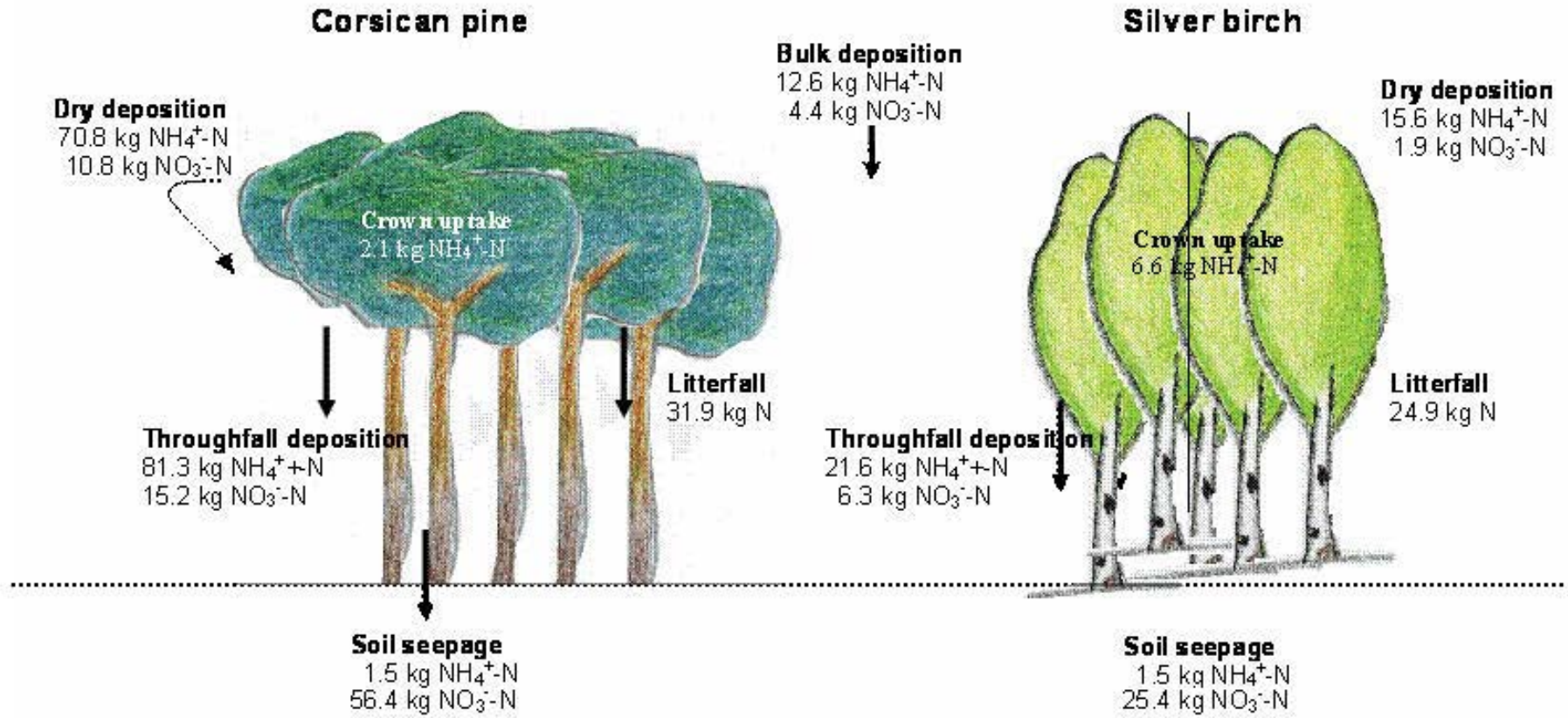

Fig. 6.4: Overview of fluxes of NH_4^+ and NO_3^- in the two forest types (at the left: Corsican pine, at the right silver birch)

On the long-term, we expect that, under comparable nitrogen loadings, nitrate seepage under Corsican pine will still increase. The C/N ratio of the forest floor, which is at the moment still 28, will probably decrease (Gundersen et al. 2006) and mineralization - so mobilization of nitrogen from the forest floor - will increase. Despite the high nitrogen deposition load, the forest floor C/N ratio is still not below the threshold value of 25 (as discussed in Chapter 5 $ 5.5.3). The seepage of nitrate probably mainly originates from nitrogen in throughfall deposition (nitrate and nitrified ammonium), and not from mineralization of organic matter. This is confirmed by Dise et al. (1998) who found higher nitrate seepage for any given forest floor C/N ratio receiving high amounts of nitrogen in throughfall. The high variability of the C/N ratios at a given level of atmospheric deposition indicates that besides deposition, other intrinsic differences in site characteristics affect the C/N ratio. We might, however, expect that when the C/N ratio of the Corsican pine plot decreases, nitrate seepage will even be higher.

6.5.7 Are the studied forest types nitrogen saturated?

We observed in both vegetation types nitrate seepage throughout the complete year so we can conclude that the availability of ammonium and nitrate is in excess of total combined plant and microbial nutritional demand.

Results from experimental studies in North America have shown that many temperate forests initially retain most of the N that is added and only respond with small seepage losses (Fenn et al. 1998, Aber et al. 1998, Johnson 1992b). After several years of N additions, however, seepage losses increase significantly (Magill et al. 1997). Many European forests receiving atmospheric N deposition behave similarly, with nitrate losses typically representing 30% of inorganic N deposition (Dise et al. 1995), although sites receiving high atmospheric N inputs for long periods have been found to approach a N input to N output ratio of 1 (Emmett et al. 1995).

6.5.8 Consequences of high nitrate seepage in both forest types

Although distinct differences in nitrate seepage were apparent in this study, both forest types showed clear symptoms of a nitrogen surplus and soil acidification processes. The forest soil is strongly impoverished because nitrate seepage is linked to losses of so-called base cations

(Carnol et al 1997a, 1997b), which reduces the buffering potential of the soil. Soil acidification is reflected in both forest types by the loss of base cations, and by the mobilisation of aluminium in particular. The seepage of NO_3^-, Ca^{2+} and Al(III) over a 1-year period was more than twice as high under Corsican pine than under silver birch. The higher seepage of anions under pine involves a higher soluble cation fraction, making total cation seepage a function of total anion seepage (Johnson 1992a).

6.6 Conclusions

The studied naturally regenerated stand of silver birch (*Betula pendula* Roth) contributes less to acidification and eutrophication of the soil than the homogeneous plantation of Corsican pine (*Pinus nigra ssp. laricio* Maire). The lower nitrogen throughfall deposition in the birch stand was reflected in a 60% lower nitrate seepage at 1 m depth compared with pine. The forest soil is strongly impoverished because nitrate seepage is linked to losses of the so-called base cations. Also higher fluxes of aluminium were reported under Corsican pine compared to silver birch, indicating a higher pollution risk for the groundwater.

7 A review of forest type effect on throughfall deposition and seepage flux

After: De Schrijver A., Geudens G., Augusto L., Staelens J., Mertens J., Wuyts K, Gielis L. & Verheyen K. A review of forest type effect on throughfall deposition and seepage flux. Oecologia, in press.

7.1 Abstract

Converting deciduous forests to coniferous plantations and *vice versa* causes environmental changes, but till now insight in the overall effect is lacking. This review, based on 38 case-studies, aims to find out how coniferous and deciduous forests differ in terms of throughfall (+ stemflow) deposition and seepage flux to groundwater. From the comparison of coniferous and deciduous stands at comparable sites, it can be inferred that deciduous forests receive less sulphur and nitrogen via throughfall (+stemflow) deposition on the forest floor. In regions with relatively low atmospheric nitrogen (N) open field deposition, lower NH_4^+ mean throughfall (+ stemflow) deposition was however reported under conifers compared to deciduous forest, while in regions with high atmospheric N pollution (>10 kg $N.ha^{-1}.yr^{-1}$), the opposite could be concluded. The higher the NH_4^+ open field deposition, the bigger the difference between the coniferous and deciduous throughfall (+ stemflow) deposition.

Furthermore, it can be concluded that canopy exchange of K^+, Ca^{2+} and Mg^{2+} is on average higher in deciduous stands. The significantly higher stand deposition flux of N and sulphur (S) in coniferous forests is reflected in a higher soil seepage flux of NO_3^-, SO_4^{2-}, K^+, Ca^{2+}, Mg^{2+} and Al(III). Considering a subset of papers for which all necessary data were available, a close relationship between throughfall (+ stemflow) deposition and seepage was found for N, irrespective of the forest type, while this was not the case for S. The higher seepage flux of anions under conifers clearly involves a higher seepage of the cations K^+, Ca^{2+}, Mg^{2+} and Al(III). This review shows that the higher input flux of N and S in coniferous forests clearly involves a higher seepage of nitrate and sulphate and accompanied cations to the groundwater, making this forest type more vulnerable to acidification and eutrophication compared to the deciduous forest type.

7.2 Introduction

Worldwide, coniferous forests are distributed far beyond their assumed natural ranges (e.g. Spiecker et al. 2004, Bustamante and Simonetti 2005). Conversion of secondary spruce or pine forests on sites where in natural conditions broadleaved deciduous species would dominate is generally agreed on in Europe (Spiecker et al. 2004). The rationale for converting coniferous forest plantations is mainly sylvicultural. For example, pure Norway spruce stands are generally considered to be highly unstable and prospects are this will persevere in the light of climate change (von Lüpke et al. 2004). The decrease in profitability of mixed stands (e.g. of Norway spruce and beech) compared to pure Norway spruce stands is compensated by the significant risk attenuation (Knoke et al. 2005). Protection of soil and water (De Schrijver et al. 2004, von Wilpert et al. 2000) and biodiversity (Gartner and Reif 2004) are further important arguments for conversion of coniferous forests.

In several cases, evergreen coniferous forests have been shown to speed up forest soil acidification (lower soil pH and lower base saturation) compared to broadleaved deciduous trees (Augusto et al. 2002). Coniferous litter is less rich in nutrients (Augusto et al. 2002, Reich et al. 2005), is more resistant to biological degradation and leaches more organic acids compared to broadleaved litter (Ranger and Nys 1994, Howard and Howard 1990, Johansson 1995). Soil biological activity and therefore nutrient turnover is reported to be lower under evergreen conifers (Binkley 1996, Saetre 1998). Furthermore, coniferous canopies scavenge atmospheric pollution more efficiently, enhancing acid deposition through higher dry deposition of SO_4^{2-}, NH_4^+ and NO_3^- (Augusto et al. 2002). The higher throughfall deposition input in coniferous forests might cause higher seepage of NO_3^-, SO_4^{2-} and accompanying cations compared to deciduous forests (Matson et al. 2002, Gundersen et al. 2006).

This information leads us to the question which environmental effect can be expected from converting coniferous forest to deciduous forest and *vice versa*. An approach to answer this question is by comparing deciduous and coniferous forest stands under comparable local site conditions. Climate (Erisman and Draaijers 2003), soil type (Silva et al. 2005, Boumans et al. 2004), land use history (Matson et al. 2002), pollutant emission level (Matson et al. 2002), succession phase (Aber et al. 1989, Agren and Bosatta 1988, Cairns and Lajtha 2005), distance to forest edge (Weathers et al. 2001), and experimental set-up (Bleeker et al. 2003) are all known to have a significant impact on the ion fluxes observed. The aim of this chapter is to find out how coniferous and deciduous broadleaved forests differ in terms of ion throughfall (+ stemflow) deposition and seepage flux to groundwater. Therefore, we analyzed

existing research results of coniferous and deciduous forests that meet the prerequisite of comparability of forest stands.

7.3 Materials and methods

7.3.1 Data collection

A literature survey was performed starting on the Web of Science (ISI Web of Knowledge v3.0 (period 1972 till October 2006)) to obtain published studies comparing values of throughfall (+ stemflow) deposition and/or seepage fluxes of ions in pairs of forest stands solely differing in tree species composition. The cited references of these studies were searched for other peer-reviewed publications. The forest stands in each pair had to be identical concerning mesoclimate, soil type, land use history, pollutant emission level and forest succession stage as well as experimental set-up.

We found 24 papers analyzing 32 useful pairs of forest stands for throughfall, and sometimes stemflow deposition flux, and 18 stand pairs for seepage flux (Table 7.1). Papers of Dambrine et al. (2000) and Jussy et al. (2004) were not selected because their experimental plots were the same as in Fichter et al. (1998), in which more ions were discussed. The paper of Wesselinck et al. (1995) was not chosen because of overlap with Heinrichs and Mayer (1977) and Matzner and Meiwes (1994), in which complementary information was given. The SB and SS plots reported in Borken et al. (2004) were the same plots as in Matzner and Meiwes (1994) so its throughfall data were not included in the statistical analysis.

Deposition in the open field (wet-only or bulk precipitation deposition in an adjacent open space) was available for the majority of the studies. Seepage flux in the soil profile was measured by different types of lysimeters at a depth varying between under the forest floor and 180 cm (Table 7.1).

Table 7.1: Field studies measuring nutrient fluxes in a coniferous and a nearby deciduous forest stand on comparable sites. OF=open field deposition, TF= throughfall deposition, SF = Stemflow deposition, S = seepage flux. For seepage flux, the depth of sampling is indicated.

	Authors	**year**	**Tree species**		**Site location**	**Soil type**	**Sampled water fractions**				**Remarks**	
			Coniferous	Deciduous			OF	TF	SF	S	Sampling depth P (cm)	
1	Augusto et al. (ref. 19)	2002	*Picea abies*	*Fagus sylvatica*	Sweden		x	x				
2	Augusto et al. (ref. 123)	2002	*Picea abies*	*Fagus sylvatica*	Germany					x	50	
3	Augusto et al. (ref 144)	2002	*Picea abies*	*Quercus petraea*	France					x	60	
4	Augusto et al. (ref. 205)	2002	*Picea abies*	*Fagus sylvatica*	Germany		x	x				
5	Bergkvist and Folkeson	1995	*Picea abies*	*Betula pendula*	Sweden	Haplic podzol	x	x	x	x	50	Plot: Munkarp
6	Bergkvist and Folkeson	1995	*Picea abies*	*Betula pendula*	Sweden	Haplic podzol	x	x	x	x	50	Plot: Nyhem
7	Borken et al. (*)	2004	*Picea abies*	*Fagus sylvatica*	Germany	Dystric cambisols		x		x		Plots SB-SS
8	Borken et al.	2004	*Pinus sylvestris*	*Fagus sylvatica*	Germany	Dystric cambisols		x		x		Plots SB-SP
9	Borken et al.	2004	*Picea abies*	*Fagus sylvatica*	Germany	Dystric cambisols		x		x		Plots UB-US
10	Cappellato et al.	1993	*Pinus taeda*	*Carya tomentosa, C. glabra, Quercus rubra, Q. alba, Liriodendron tulpifera*	USA	Inceptisol/ Ultisols	x	x				
11	De Schrijver et al.	2004	*Pinus nigra*	*Betula pendula*	Belgium	Haplic podzol	x	x		x	100	
12	Durand et al.	1992	*Picea abies*	*Fagus sylvatica*	France	Acid coarse-textured organic soil type	x	x				
13	Fichter et al.	1998	*Picea abies*	*Fagus sylvatica*	France	Acid distrochrept soil	x	x		x	60	
14	Heinrichs and Mayer	1977	*Picea abies*	*Fagus sylvatica*	Germany	Acid Braunerde	x	x	x	x	50	
15	Herman et al.	2006	*Pinus sylvestris*	*Betula pendula, Quercus robur*	Germany	Humo-ferric podzols	x					Plots P2-OB3
16	Houle et al.	1999	*Abies balsamea, Picea rubens*	*Acer saccharum*	Canada	Ferro-humic podzols	x	x	x			
17	Künstle et al.	1981	*Picea abies*	*Fraxinus excelsior, Acer sp.*	Germany		x	x				pair 1
18	Künstle et al.	1981	*Pseudotsuga menziesii*	*Fagus sylvatica*	Germany		x	x				pair 2

Table 7.1: Continued

	Authors	year	Tree species		Site location	Soil type	Sampled water fractions				Remarks	
			Coniferous	Deciduous			OF	TF	SF	S	Sampling depth (cm)	
19	Künstle et al.	1981	*Picea abies*	*Fagus sylvatica*	Germany		x	x				pair 3
20	Künstle et al.	1981	*Picea abies*	*Fagus sylvatica*	Germany		x	x				pair 4
21	Lochman and Sebkova	1998	*Picea abies*	*Betula pendula*	Czech Republic	*Dystric Gleyic Cambisol*	x	x	x		?	data from Table I
22	Lochman et al.	2004	*Picea abies*	*Fagus sylvatica*	Czech Republic	*Distric Cambisol*	x	x				pair plot III and plot V
23	Matzner and Meiwes	1994	*Picea abies*	*Fagus sylvatica*	Germany	Typic Distrochrept	x	x				
24	McDowell et al.	2004	*Pinus resinosa*	*Quercus velutina, Quercus rubra*	USA	Typic Distrochrept				x	Under forest floor	treatment of no N addition
25	McDowell et al.	2004	*Pinus resinosa*	*Quercus velutina, Quercus rubra*	USA	Typic Distrochrept				x	Under forest floor	treatment of low N addition
26	McDowell et al.	2004	*Pinus resinosa*	*Quercus velutina, Quercus rubra*	USA	Typic Distrochrept				x	Under forest floor	treatment of high N addition
27	Neary and Gizyn	1994	*Pinus strobus, Tsuga canadensis, Acer rubrum, Quercus rubra*	*Acer saccharum, A. rubrum, Fagus grandifolia, Betula alleghaniensis, B. papyrifera, Populus grandidentata*	Canada	Podzol	x	x	x			
28	Nihlgard	1970	*Picea abies*	*Fagus sylvatica*	Sweden	Acid brown forest soil	x	x	x			
29	Pucket	1990	*Pinus strobus*	*Quercus prinus, Nyssa sylvatica*	USA		x	x				
30	Ranger and Nys	1994	*Picea abies*	*Fagus sylvatica, Quercus petraea*	France	*Typic Distrochrept*	x	x		x	45	
31	Rothe et al.	2002	*Picea abies*	*Fagus sylvatica*	Germany	*Distric Cambisol*	x	x		x	175	mean of pair 1 and 2
32	Rothe et al.	2002	*Picea abies*	*Fagus sylvatica*	Germany	*Distric Cambisol*	x	x	x	x	100	pair 3
33	Shibata and Sakuma	1996	*Pinus strobus, Pinus koraensis*	*Quercus mongolica, Magnolia obovata, Fraxinus lanuginosa, Rhus trichocarpa*	Japan	Volcanogenous Regosols	x	x				data from Table 3

Table 7.1: Continued

	Authors	**year**	**Tree species**		**Site location**	**Soil type**	**Sampled water fractions**				**Remarks**	
			Coniferous	Deciduous			OF	TF	SF	S	Sampling depth (cm)	
34	Singer et al.	1996	*Pinus halepensis*	*Quercus calliprinos*	Israel	Calcareous soil type	x	x	x			
35	Van Ek and Draaijers	1994	*Pinus sylvestris*	*Quercus robur*	The Netherlands	Sandy soil type	x	x				
36	Van Ek and Draaijers	1994	*Pseudotsuga menziesii*	*Quercus robur*	The Netherlands	Sandy soil type	x	x				
37	Von Wilpert et al. (**)	2000	*Picea abies*	*Fagus sylvatica*	Germany	*Cambisol*	x	x	x	x	180	pair 1 WF2 and WBu
38	Von Wilpert et al. (**)	2000	*Picea abies*	*Fagus sylvatica, Abies alba, Picea abies*	Germany	*Cambisol*	x	x	x	x	180	pair 2 WF4 and Misch

(*) throughfall collection was performed in the same plots as Matzner and Meiwes (1994), so excluded from statistical analysis ,

(**) datasheets received from von Wilpert (2004

7.3.2 Data analysis

Compared to precipitation in the open field, the chemical composition of throughfall and stemflow water is generally altered, and it is widely acknowledged that this transformation results from (i) dry deposition of aerosols and gases on leaves, branches and stems, as well as (ii) canopy exchange processes through diffusion and/or exchange between the water layer covering leaves/needles and the underlying apoplast and uptake of gases through stomata (Draaijers et al. 1997). In order to distinguish internal and external input sources of K^+, Ca^{2+} and Mg^{2+} to the ecosystems, we used the so-called filtering approach described in the canopy budget method of Ulrich (1983d). In this method, Na^+ is used as tracer ion to calculate dry deposition of K^+, Ca^{2+} and Mg^{2+}, since Na^+ is assumed to be inert with respect to the canopy, *i.e.* neither uptake nor leaching occurs, and particles containing K^+, Ca^{2+} and Mg^{2+} are assumed to have the same deposition velocity as those containing Na^+. This analysis could be performed for 17 out of the 33 plots for which throughfall and bulk deposition was reported. For more details on the method, see §5.3.4.

To compare the results of the different studies, a simple meta-analysis was performed, considering Gurevitch et al. (2001). Because of the lack of reporting on sample standard deviation in most studies, the followed approach is less powerful and less accurate than the standard meta-analysis statistical procedures (Gurevitch et al. (2001)). Data of ion fluxes measured in different years and reported separately were not considered as repetitions but were averaged. The chosen metric of effect size was the ratio between the ion flux in the coniferous forest stand and the ion flux in the deciduous forest stand. These ratios were calculated for (1) throughfall + stemflow deposition, (2) dry deposition factor, and for (3) seepage fluxes. Mean ratios were calculated for each considered ion, and a 95% bootstrap confidence interval of this mean was calculated. Ion fluxes differ significantly between the coniferous and deciduous forest stands when the 95% bootstrap confidence interval of the mean ratio does not include 1. This procedure was followed for ions with at least 8 repetitions. The following ions for deposition and seepage fluxes were reported in the studies considered: NH_4^+, NO_3^-, sum of NH_4^+ and NO_3^-, SO_4^{2-}, Na^+, K^+, Ca^{2+}, and Mg^{2+}. Too few data pairs were available for Al(III) and HCO_3^- deposition fluxes and H^+ seepage flux to draw meaningful conclusions.

The level of atmospheric pollution differed much between the studies. Especially forest plots in heavily urbanized regions, near industrial plants or near intensive animal breeding farms

received much more N deposition than sites in remote, semi-natural areas. To forest ecosystems, an open field deposition level of 10 kg $ha^{-1}.yr^{-1}$ (714 $mol.ha^{-1}.yr^{-1}$) of total inorganic N (NH_4^+-N + NO_3^--N) seems to be a threshold above which N cycling changes (Kristensen et al. 2004). Therefore, besides analyzing the global dataset, a distinction was made between stand pairs with a low (<10 $kgN.ha^{-1}.yr^{-1}$) and a high inorganic N deposition in the open field (>10 kg N ha^{-1} yr^{-1}).

The relation between the bulk deposition in the open field and the ratio of throughfall (+ stemflow) deposition under coniferous and deciduous canopies was explored for NH_4^+, NO_3^-, SO_4^{2-} and Na^+, K^+, Ca^{2+} and Mg^{2+} through linear regression between the log-transferred data.

7.4 Results

7.4.1 Throughfall (+ stemflow) deposition flux

Considering all available data, the ratio of throughfall (+ stemflow) deposition flux under coniferous and deciduous canopies is higher than one for both NH_4^+ and NO_3^-, as is for their sum (Table 7.2). The mean ratio equals to 1.72 for both NO_3^- and NH_4^+. Distinguishing forest pairs with a low (≤ 714 $mol.ha^{-1}.yr^{-1}$) from a high inorganic N deposition in the open field (≥ 714 $mol.ha^{-1}$ yr^{-1}) reveals that in the low deposition plots mean stand deposition flux of NH_4^+ is lower in the coniferous stands, although not significantly, while NO_3^- is higher in the coniferous forest type. On the contrary, for the stand pairs with high open field N deposition, the mean stand deposition flux of both NH_4^+ and NO_3^- is higher in the coniferous stands (mean ratios = 2.01 and 1.85, respectively).

SO_4^{2-} throughfall (+ stemflow) deposition is higher under coniferous canopies than under deciduous canopies (mean ratio = 1.74) (Table 7.2). H^+ throughfall (+ stemflow) deposition is on average 2.2 times higher in coniferous stands as compared to deciduous stands, but the mean ratio is very variable (confidence interval 1.56-2.66). The mean ratio of throughfall (+ stemflow) deposition fluxes under coniferous and deciduous canopy is higher than one for all the so-called base cations, except for K^+. The mean ratio for Na^+ is highest of the base cations, followed by Ca^{2+}, Mg^{2+} and K^+ in decreasing order of magnitude.

Since the coniferous/deciduous combination *Picea abies*/*Fagus sylvatica* is frequently abundant in our dataset (16 of the 38 studies), we calculated the ratio of throughfall (+ stemflow) deposition flux for stand comparisons other than *Picea abies*/*Fagus sylvatica*

(Table 7.2). We found highly comparable values for both the mean ratio and bootstrap confidence intervals for all considered ions other than Ca^{2+} and Mg^{2+}.

For a subset of 17 plots, the dry deposition factor (DDF) was calculated. The DDF is higher for coniferous than for deciduous forest types and its ratio ranges from 2.2 to 5.7. Canopy exchange values are highest for the deciduous forest type and are on average 428 $eq.ha^{-1}.yr^{-1}$ for K^+, 208 $eq.ha^{-1}.yr^{-1}$ for Ca^{2+} and 116 $eq.ha^{-1}.yr^{-1}$ for Mg^{2+}, while for the coniferous sites mean values range from 348 $eq.ha^{-1}.yr^{-1}$ for K^+ over 165 $eq.ha^{-1}.yr^{-1}$ for Ca^{2+} and 70 $eq.ha^{-1}.yr^{-1}$ for Mg^{2+} (Fig. 7.1). For Ca^{2+}, negative values for canopy exchange were calculated for the coniferous plots 11, 15 and 33 and for the deciduous plot 12 and for Mg^{2+} for the coniferous and deciduous plots 12 and 14.

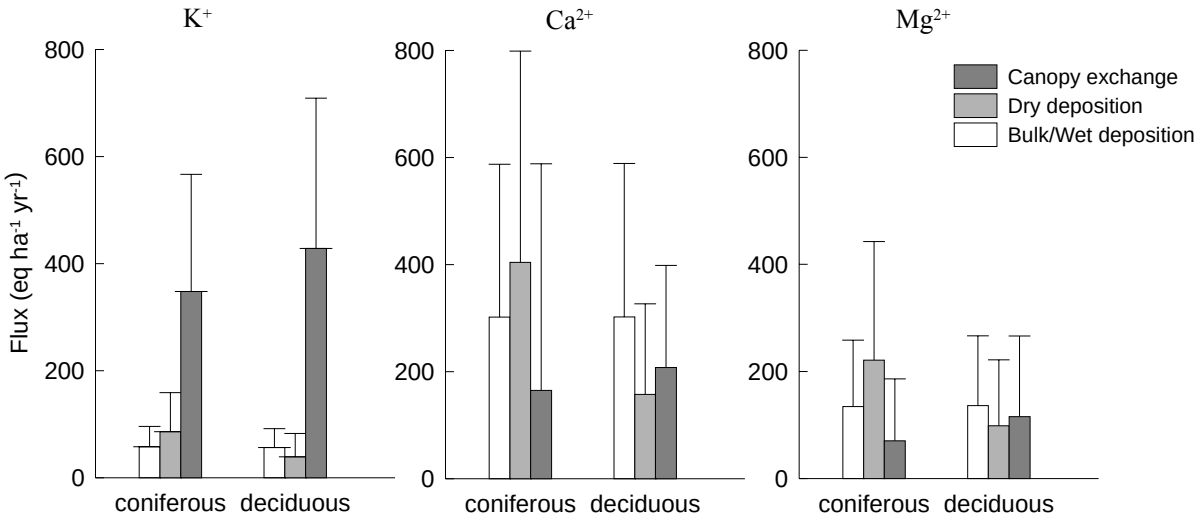

Fig. 7.1: Average values ($eq.ha^{-1}.yr^{-1}$) and standard deviation of bulk deposition, dry deposition and canopy exchange of K^+, Ca^{2+} and Mg^{2+} for coniferous and deciduous plots

Excluding the *Picea abies*/*Fagus sylvatica* combinations from our dataset, we obtained a nearly identical mean ratio value for the dry deposition factor (DDF). The bootstrap confidence interval ranges between 1.87 and 6.78. The canopy exchange values for Ca^{2+} and Mg^{2+} calculated for four subsets (*Picea abies*, *Fagus sylvatica,* other conifers than *Picea abies*, and other deciduous species than *Fagus sylvatica*) reveal that the deciduous species other than *Fagus sylvatica* have a higher mean Ca^{2+} and Mg^{2+} canopy exchange value compared to the other three subsets (numbers not shown). *Fagus sylvatica* shows for all considered cations the lowest mean canopy exchange.

For the ions K^+, Ca^{2+}, Mg^{2+} and SO_4^{2-}, no significant relationship was found between the open field deposition and the ratio of throughfall (+ stemflow) deposition under coniferous and deciduous canopies. The log-transformed ratios of Na^+ and NO_3^- plotted against their respectively log-transformed open field depositions show a significant but rather weak positive relationship (Fig. 7.2 with log-transformed data, for Na^+: y=0.091.x-0.05; p = 0.020, R^2_{adj} = 0.251; for NO_3^-: y=0.22.x-0.32; p = 0.043, R^2_{adj} = 0.175). For NH_4^+, however, this linear regression was found to be quite strong and significant (y=0.63.x-1.48; p = 0.001, R^2_{adj} = 0.481) (Fig. 7.2).

7.4.2 Seepage flux

The ratio of ion seepage under coniferous and deciduous stands differed from one ($p < 0.05$) for all ions considered (Table 7.3). Seepage of NO_3^- was found to be on average almost 12 times higher under coniferous forest stands than under deciduous forest stands. We could not distinguish between regions with low and high atmospheric N pollution level, because the number of replicates would become too low for a meaningful analysis. Seepage of SO_4^{2-} was found to be on average 1.64 times higher under coniferous compared to deciduous stands. Significant differences between the two stand types were also found for Al(III) and K^+, Ca^{2+} and Mg^{2+}. The mean ratio and the range of the confidence interval was found to be highest for aluminium seepage (4.46). Because of the relatively low number of studies, we did not distinguish between plot combinations with or without *Picea abies*/*Fagus sylvatica.*

A quite strong relationship (R^2_{adj} = 0.619, $p<0.001$) was found, irrespective of the forest type, between the log-transformed values for the sum of seepage of the cations K^+, Ca^{2+}, Mg^{2+} and Al(III) and the seepage of SO_4^{2-} (Fig 7.3a) (Equation 7.1).

$$\log(seepage_{(K^+ + Ca^{2+} + Mg^{2+} + Al(III))}) = -1.044 + 1.43 \times \log(seepage_{SO_4^{2-}}) \qquad (7.1)$$

No significant relationship was found between seepage of the mentioned cations and seepage of NO_3^-.

Table 7.2: Mean ratio between ion stand (throughfall + stemflow) deposition fluxes under coniferous forest and under deciduous forest. The 95% bootstrap confidence interval is given between brackets. N = number of studies. Results are given for: (1) all available data, (2) data with open field inorganic N deposition flux ≤ 714 mol.ha^{-1}.yr^{-1} and (3) > 714 mol.ha^{-1}.yr^{-1}

	All data			**Data other than *Picea/Fagus*-comparison**			**Data $N_{open\ field}$ < 714 mol ha^{-1} yr^{-1}**			**Data $N_{open\ field}$ ≥ 714 mol ha^{-1} yr^{-1}**		
Ion	N	Mean		N	Mean		N	Mean		N	Mean	
NH_4^+	18	1.72	[1.47-2.08]	12	1.64	[1.25-2.08]	5	0.81	[0.70-1.07]	13	2.01	[1.81-2.43]
NO_3-	19	1.72	[1.58-1.86]	13	1.66	[1.45-1.88]	6	1.35	[1.09-1.61]	13	1.85	[1.72-2.02]
NH_4^++NO_3^-	22	1.85	[1.64-2.07]	11	1.74	[1.39-2.13]	6	1.28	[0.97-1.83]	16	2.03	[1.81-2.26]
H^+	12	2.22	[1.56-2.66]	8	2.22	[1.50-2.97]						
SO_4^{2-}	28	1.74	[1.57-1.92]	13	1.66	[1.38-1.95]						
Na^+	18	1.45	[1.36-1.54]	11	1.47	[1.32-1.61]						
K^+	24	1.09	[0.93-1.28]	11	0.81	[0.56-1.10]						
Ca^{2+}	25	1.37	[1.25-1.50]	12	1.14	[0.98-1.32]						
Mg^{2+}	18	1.22	[1.08-1.35]	11	1.07	[0.92-1.25]						
DDF	17	3.73	[2.24-5.68]	11	3.74	[1.87-6.78]						

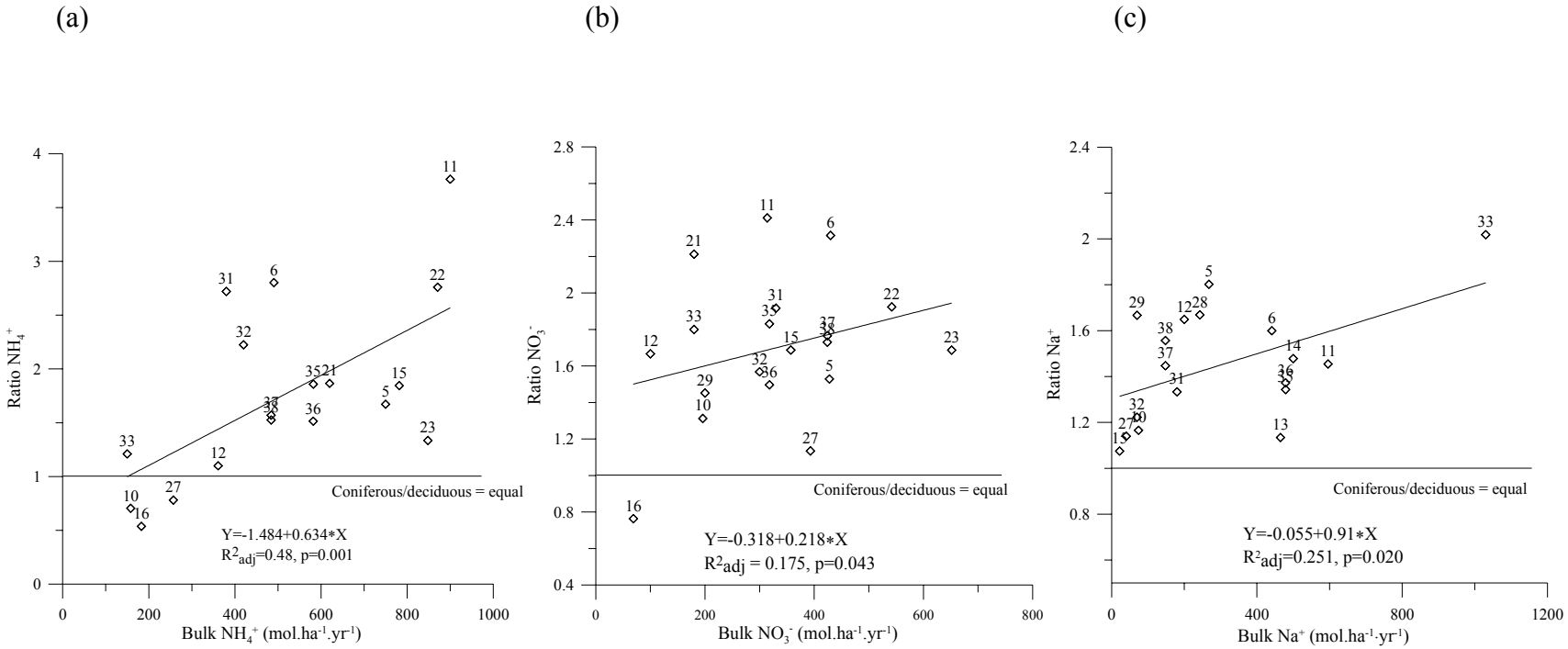

Fig. 7.2: Scatter plot of the ratio of the throughfall (+stemflow) deposition flux of (a) NH_4^+, (b) NO_3^- and (c) Na^+ in coniferous paired deciduous stands for its increasing yearly deposition fluxes in open field precipitation adjacent to each stand pair. The labels refer to the index numbers of the studies in Table 7.1.

Table 7.3: Mean ratio between ion fluxes in seepage water under coniferous forest and under deciduous forest. N = number of studies. The 95% bootstrap confidence interval is given between brackets

Ion	N	Mean	
NH_4^+	13	1.30	[1.02-1.57]
NO_3-	13	11.98	[4.7-21.01]
NH_4^++NO_3^-	18	10.34	[4.09-13.24]
SO_4^{2-}	12	1.64	[1.23-2.06]
Na^+	10	1.41	[1.04-1.83]
K^+	12	1.94	[1.50-2.43]
Ca^{2+}	12	1.81	[1.31-2.42]
Mg^{2+}	12	1.91	[1.30-2.11]
Al (III)	8	4.46	[1.31-9.66]

7.4.3 Relationship throughfall (+stemflow) deposition and seepage flux

For a subset of plots for which all necessary data were available, a strong relationship was found ($n = 20$, $R^2_{adj} = 0.632$, $p < 0.001$) between the log-transformed throughfall (+ stemflow) deposition and the log-transformed seepage flux of total inorganic nitrogen ($NO_3^- + NH_4^+$) (Fig 7.3b with non log-transformed values), while for SO_4^{2-} (Fig 7.3c with non log-transformed values) this was not the case ($n = 18$, $R^2_{adj} = -0.057$, $p = 0.767$). No significant linear regression could be fitted between the log-transformed values of throughfall (+ stemflow) depositions of N and S and the sum of seepage of K^+, Ca^{2+}, Mg^{2+} and Al (III).

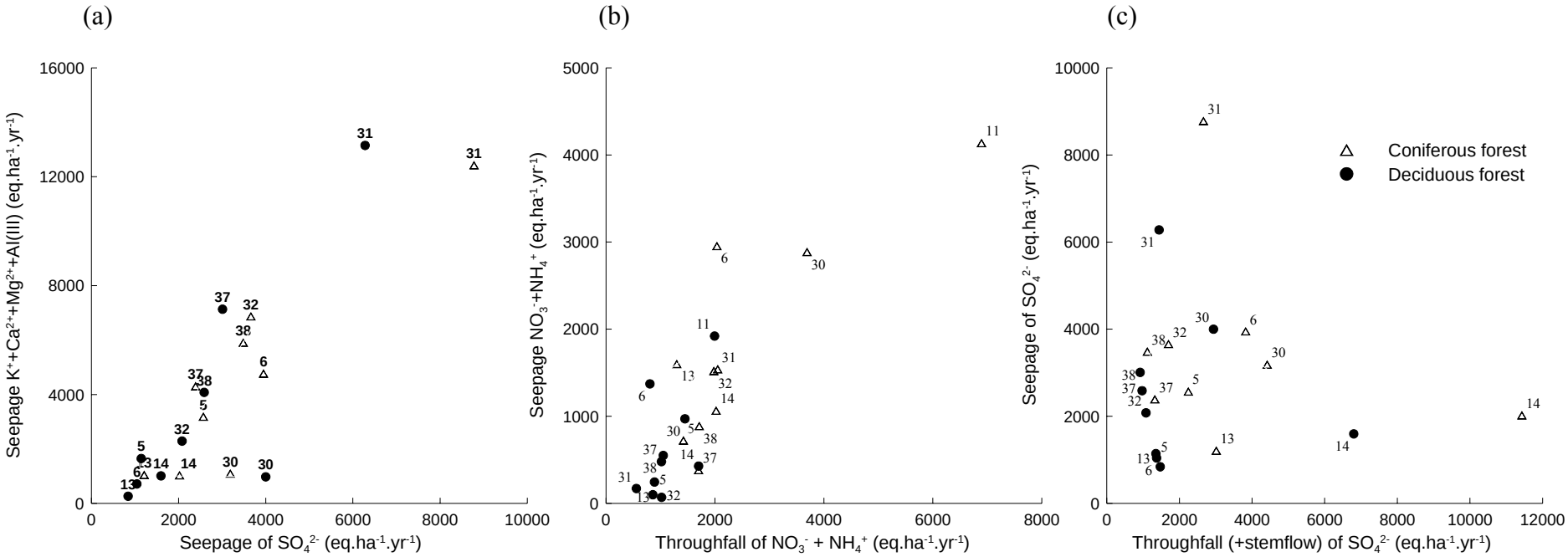

Fig 7.3: Relationship between (a) seepage fluxes of K^+, Ca^{2+}, Mg^{2+} and Al(III) and of NO_3^- and SO_4^{2-}, throughfall (+stemflow) deposition flux and seepage of (b) total inorganic N and (c) SO_4^2. The labels refer to the index numbers of the studies in Table 7.1.

7.5 Discussion

7.5.1 Do coniferous and deciduous forest types differ in their throughfall deposition flux?

Coniferous forests annually intercept more atmospheric pollutants than deciduous forests at the same site and under the same climatic conditions. This can be concluded from the analysis of the calculated dry deposition factor of 17 stand pairs. Important causal factors are differences in vegetation structure, such as the generally lower height and LAI in deciduous stands (Augusto et al. 2002), a higher stand and crown density and volume (Cole and Rapp 1981) and the summergreen character of deciduous stands (Alcock and Morton 1985, Draaijers 1993, Houle et al. 1999, Erisman and Draaijers 2003). In addition, the leaf shape affects the amount of ions deposited. Leaves with long narrow shape (needles) are according to Woodcock (1953 in Smith 1981) more efficient in salt accumulation than circular ones. Nevertheless, during the growing season, higher dry deposition fluxes to deciduous forests in comparison to coniferous forests have been observed (Cappellato et al. 1993).

The higher interception capacity can explain the significantly higher throughfall (+ stemflow) deposition of nitrogen and sulphur in coniferous forests. From the analysis of Na^+ in Fig. 7., which can be used as a tracer ion to get insight in the dry deposition capacity, it can be carefully concluded that the higher the deposition load, the greater the difference in filtering capacity between coniferous and deciduous forests. So in regions with higher concentrations of nitrogen in the atmosphere, it can be expected that coniferous forests filter more efficiently. The higher the ammonium open field deposition, the bigger indeed the difference between its coniferous and deciduous throughfall (+ stemflow) deposition is. However, in regions with relatively low atmospheric N open field deposition ($\leq$ 714 mol N $ha^{-1}.yr^{-1}$), lower NH_4^+ mean throughfall (+ stemflow) deposition is reported under conifers compared to deciduous forests in three of the five plots. In regions with high atmospheric N pollution, coniferous throughfall (+ stemflow) deposition of nitrogen is in all considered plots higher than in deciduous plots (on average about 2 times). We hypothesize that, when open field N deposition is low, higher yearly canopy uptake of NH_4^+ by coniferous forests may keep pace with the higher dry deposition as compared to deciduous forests, resulting in the observed lower NH_4^+ stand deposition flux under conifers in part of the plots. In regions with high atmospheric N pollution, the canopy uptake of NH_4^+ may be obscured by elevated dry deposition, with a higher stand deposition flux of NH_4^+ in coniferous forests as a result. This hypothesis is very theoretical considering the small number of repetitions in the low deposition region, and more research is needed to confirm it. Since NH_4^+ is taken up at a significantly faster rate than NO_3^-

(Bowden et al. 1989, Boyce et al. 1996, Garten and Hanson 1990, Schulze and Gebauer 1989, Stachurski and Zimka 2000), a less clear compensation process is observed for NO_3^- and the higher dry deposition of NO_3^- onto coniferous forests therefore usually results in higher stand deposition fluxes, also in regions with lower atmospheric N pollution. Only for ref. 16 in Table 7.1 (Houle et al. 1999), a lower NO_3^- throughfall deposition flux was measured under coniferous forest compared to deciduous forest.

The higher throughfall (+ stemflow) proton input in the coniferous forests is again an indication of their higher dry deposition capacity. In addition, the higher proton input in the coniferous forests can be due to the canopy exchange of NH_4^+ for H^+ (Neary and Gizyn 1994), as coniferous forests probably retain more NH_4^+ in the canopy. But also exchange processes of H^+ for so-called base cations in the crown are of significant influence on the throughfall (+ stemflow) deposition flux of protons (Ulrich 1983), and consequently on the acidity of throughfall and stemflow water.

Opposite to Na^+, throughfall (+ stemflow) deposition fluxes of K^+, Ca^{2+} and Mg^{2+} are highly affected by canopy exchange processes. From our calculations it can be concluded that canopy exchange of these cations is on average higher in the deciduous stands. The variability on the calculations is high, which is attributable to the high diversity in species, ages, and stand characteristics, all factors of significant influence on canopy exchange processes. For both forest types, K^+ is the most important cation involved in the exchange process (K^+ exchange contributes on average 60% of the total exchange of K^+, Ca^{2+} and Mg^{2+} for coniferous forests and 57% for deciduous forests), followed by Ca^{2+} (28% for both forest types) and Mg^{2+} (12 and 15%, respectively). For Ca^{2+} and Mg^{2+}, negative canopy exchange values were obtained for a small number of deciduous and coniferous stands. This could indicate canopy uptake instead of leaching, but most probably is a bias on the filtering approach. In contrast to Ca^{2+} and Mg^{2+}, no significant differences between the forest types are found for K^+ throughfall (+ stemflow) deposition, which may indicate that the higher dry deposition from the atmosphere (soil dust and sea salt) to coniferous forest stands compensates for the higher canopy exchange in the deciduous forest stands (Alcock and Morton 1985, Houle et al. 1999, Van Ek and Draaijers 1994). Dry deposition is the major source of Ca^{2+} and Mg^{2+} in coniferous forests, while in deciduous forests open field deposition dominates the composition of throughfall (+ stemflow) water.

The effect of forest type on canopy exchange processes can be direct or indirect. Direct impact can be caused by differences in canopy characteristics such as aboveground biomass,

nutrient amount, species specific concentrations in leaves (Lovett et al. 1989) and evergreen character (Smith 1981). Nutrient concentrations in leaves are higher compared to needles (Johansson 1995, Reich et al. 2005). The type of forest can also indirectly influence canopy exchange through forest soil characteristics such as extractable amount of cations in the soil solution (Lovett and Schaefer 1992).

7.5.2 Do coniferous and deciduous forest types differ in their seepage flux?

The significantly higher throughfall (+ stemflow) deposition flux of inorganic N in coniferous forests is clearly reflected in a higher soil seepage flux of NO_3^-. In all stand comparisons included in the meta-analysis except one (von Wilpert et al. 2000, ref. 37 in Table 7.1), NO_3^- seepage under coniferous forests is higher than under deciduous forests, although the differences between forest types are highly variable. In the case-study of von Wilpert et al. (2000), NO_3^- seepage at a depth of 40 cm was nevertheless higher under the *Picea abies* stand compared to the *Fagus sylvatica* stand. The opposite conclusion at 180 cm depth could be possibly due to higher denitrification under *Picea abies* (von Wilpert, personal communication).

For a subset of data we found a close relationship between N throughfall (+ stemflow) deposition and N seepage and this irrespective of forest type. Several other studies demonstrated a relationship between the level of N deposition and level of N seepage, at least for those sites where N saturation is evident (Matson et al. 2002, Macdonald et al. 2002, Gundersen et al. 1998). This brings us to the conclusion that throughfall N deposition might be the dominant trigger of NO_3^- seepage flux to groundwater.

The higher deposition load of SO_4^{2-} in coniferous forest stands was also reflected in a significantly higher seepage to groundwater. The differences between coniferous and deciduous forests in seepage flux are not that variable as for nitrate, and three out of the 11 studies report higher SO_4^{2-} seepage under deciduous forests. Other than for NO_3^-, no significant relationship between throughfall (+ stemflow) deposition and seepage flux was found for SO_4^{2-}. The ability of many soils to retain significant quantities of SO_4^{2-} by inorganic adsorption mechanisms (Harrison and Johnson 1992, Martinson et al. 2005) may underlie this conclusion. E.g. adsorption typically increases as the soil pH and the amount of organic matter decrease (Harrison and Johnson 1992), both parameters being influenced by forest type.

We found a close relationship between seepage of SO_4^{2-} and NO_3^- on the one hand, and K^+, Ca^{2+}, Mg^{2+} and Al(III) on the other hand. The higher seepage flux of anions under conifers clearly involves a higher soluble cation fraction, making total cation seepage a function of total anion seepage (Johnson 1992). Due to the lack of enough replicates, we did not distinguish between plots with low and high soil pH and base saturation. In literature it can be found that in forest soils with a base saturation over 25% or a pH-KCl value over 4.5, a strong positive relationship exists between concentrations of Ca^{2+} and strong acid anions in the subsoil (de Vries et al. 2003), indicating that the seepage of anions in less acidified soils is mainly neutralized by the release of the cations K^+, Ca^{2+} and Mg^{2+}. In forest soils with a base saturation below 25% or a pH-KCl value below 4.5, the concentration of Al(III) in the subsoil is strongly related to the concentration of SO_4^{2-} and NO_3^- (de Vries et al. 2003). Also Rothe et al. (2002) found aluminium release to be the dominant buffering process in acid forest soils.

7.5.3 Robustness of the present analysis and recommendations for future research

Approximately 40% of the studies included in this meta-analysis (16 of the 38 case studies) comprise comparisons of *Picea abies* with *Fagus sylvatica*. Our analysis shows that an exclusion of these paired comparisons does not influence the results of the meta-analysis for throughfall (+ stemflow) deposition of NH_4^+, NO_3^- and their sum, SO_4^{2-}, H^+ and Na^+. This illustrates that the results considering acidifying and N deposition are robust and can be generalized to other coniferous evergreen and deciduous broadleaved comparisons. For Ca^{2+} and Mg^{2+}, the results of the meta-analysis after exclusion of the *Picea abies*/*Fagus sylvatica* combinations are different from the complete dataset, which is probably attributable to the low canopy exchange quantities of *Fagus sylvatica* compared to other deciduous tree species. More research is needed to confirm these results and its possible interaction with soil fertility. For seepage fluxes, the relatively low number of case studies did not allow to study the impact of the high frequency of *Picea abies*/*Fagus sylvatica* combinations.

To adequately answer how coniferous and deciduous forest types differ in terms of throughfall (+ stemflow) deposition and seepage flux, we were very rigorous in selecting case studies with similar conditions for both forest types. Therefore we only selected peer-reviewed papers on the Web of Science (period 1972 - September 2006) which fulfilled the conditions of comparability described in the Material and Methods. The collected dataset is therefore not very extensive, which indicates the need for further research. The quantitative and qualitative comparability of forest ecosystem nutrient flux studies would be improved

when results would report yearly fluxes as a mean with standard deviation of at least three replicate plots per ecosystem type for each measuring year.

7.6 Conclusions

From the comparison of pure coniferous and deciduous forests at comparable sites, it can be inferred that deciduous forests receive less sulphur and nitrogen via throughfall (+ stemflow) deposition on the forest floor. This is caused by the higher dry deposition capacity of coniferous trees. Although the throughfall deposition input of K^+, Ca^{2+} and Mg^{2+} was on average higher in coniferous forests, canopy exchange of these cations was highest in deciduous forests. As a consequence of the higher throughfall input, seepage quantities of nitrogen, sulphate and cations to the deeper soil and groundwater is significantly higher under coniferous forests compared to deciduous forests. Furthermore, we found a close relationship between nitrogen throughfall (+ stemflow) deposition and seepage quantities, irrespective of the forest type, while this was not the case for sulphate.

8 Summarizing conclusions

In this chapter answers are presented to the questions formulated in chapter 1 (see §1.3). These answers provide at the same time a summary of the main aims and findings of this work. Furthermore, a discussion is presented about the risks and consequences of the observed acidification and eutrophication in forest ecosystems on sandy soil, also in relation to forest ecosystem recovery.

8.1 Are sandy forested podzols acidified in Flanders, to what extent and what are the possible causes?

Sandy forested podzols did significantly acidify over the period 1950-2000. This could be concluded from a repeated survey (in 2000, 1985, and 1950) of 29 Flemish forest ecosystems on sandy soil, of which 22 coniferous and 7 deciduous forests. We found significant differences in soil pH-KCl (which is a measure for potential acidity of the soil) between the measured years and this over the whole studied soil profile (0-50 cm depth). While the pH values dropped between 1950 and 1985 over the complete soil profile, changes between 1985 and 2000 were more pronounced and significant in the upper 10 cm of the mineral soil. The average annual acidification rate of this upper horizon was significantly higher in the period 1985-2000 than in the period 1950-1985, while in the deeper horizons no significant changes in acidification rate were found. Also in the forest floor (A_0 horizon) significant pH changes were noted.

While during the period 1950-1985 mineral soils were still in the cation exchange buffer range (for explanation on buffer ranges see §1.2.2), in 2000 the upper 10 cm had already evolved towards the aluminium buffer range. In this buffer range, Al^{3+} becomes the dominant cation in the soil solution and can be leached with seepage water. Concentrations of Al^{3+} might become toxic for plant roots (see §1.2.2 and §8.7).

Possible causes for soil acidification comprise natural soil forming processes, forest growth and anthropogenic activities such as the input of potentially acidifying deposition. We

compared Flemish and European literature data on possible causes for proton inputs and hypothesized that potentially acidifying deposition probably was the main cause for soil acidification in the observed forest soils. The relation between soil acidification and atmospheric deposition load was further explored in Chapter 3 and summarized in §8.2.

The fact that S deposition was the dominant potentially acidifying pollutant can explain why the average pH dropped in the complete soil profile during the period 1950/67-1985, since neutralisation of sulphate occurs throughout the whole soil profile (Ulrich 1983b). After 1985, acid sulphur (S) deposition decreased with more than 50% in northern Belgium. In contrast, potentially acidifying ammonium deposition almost doubled between 1950 and 1980, which may explain why soil acidification between 1985 and 2000 has been mainly restricted to the upper soil horizons. When NH_4^+ is deposited, it can be taken up by plant roots (generating 1 H^+), assimilated into organic matter (+1H^+), fixed at the cation exchange complex (CEC) (1H^+) or oxidized to nitrate (nitrification, +2H^+). All these reactions are generating protons and mainly occur in the upper soil horizons, since NH_4^+ is not mobile at all. This is confirmed by the low amounts of NH_4^+ detected in the soil solution at soil horizons deeper than 10 cm.

Supposing (although in reality not valid) that all potentially acidifying input is buffered in the forest floor and upper 10 cm of the mineral soil, we calculated that only 205 to 276 years of buffering capacity are still left. In reality, this period will obviously be longer than calculated, since part of the acid is buffered in deeper horizons, for which no data of acid neutralizing capacity (ANC) are available. These numbers clearly give an indication that soil buffering is not an endlessly continuing process.

8.2 Does atmospheric deposition significantly contribute to soil acidification?

It is often very difficult to isolate the role of anthropogenic versus natural sources of acidity in the processes affecting soil acidification. We found a significant relationship between NH_4^+ throughfall deposition and soil pH-H_2O in a homogeneous Corsican pine forest along a transect of 1.4 km length. The forest was surrounded by intensive live stock breeding that was responsible for considerable emissions of NH_3. The atmospheric dry deposition of NH_x onto this pine forest was very high due to the proximity of the sources of NH_x in the near surrounding, but also because of the higher filtering capacity of coniferous tree canopies in comparison to other vegetation types. At forest edges, dry deposition is even more elevated since the vertical wind profile is disturbed causing higher air turbulences.

The significant range in magnitude of ammonium throughfall deposition (and probably also of sulphate and nitrate) along the transect offered the opportunity to study its impact on soil acidity under comparable site and climatic conditions. We can assume that acidification as a consequence of organic acid deprotonation and forest growth is quite homogeneous along the transect. At both forest edges, the higher atmospheric potentially acidifying deposition was reflected in a significantly lower pH of the topsoil.

The topsoil was very acid along the complete length of the transect: pH-H_2O, which is a measure for actual acidity, varied from 3.05 (edge) to 3.8 (centre), while pH-KCl values varied from 2.35 (edge) to 2.8 (centre). In the forest interior, soils were in the aluminium buffer range, while at the forest borders soils already moved on to the iron buffer range, meaning that proton buffering ceases and Fe^{2+} ions attain measurable concentrations in the soil solution.

NH_4^+ can play an important role as potentially acidifying component, not only because of its high throughfall deposition flux in itself, but also by enhancing the deposition of sulphate and probably also of nitrate due to the so-called process of co-deposition. In an ICP monitoring plot, located nearby the transect, we found a significant correlation between throughfall concentrations of NH_4^+ at the one hand and of NO_3^- and SO_4^{2-} at the other hand. This indicates that besides NH_4^+ throughfall deposition, also NO_3^- and SO_4^{2-} are probably elevated at the forest edges.

8.3 How vulnerable to acidification and eutrophication are Flemish forest soils?

The concept of critical load (CL) was defined to express the tolerance of natural and semi-natural ecosystems for anthropogenic air pollution (Nillson & Grennfelt 1988). It relates to the long-term maximum exposure to one or more pollutants without occurrence of any significant harmful effect on for example biodiversity, forest vitality or nitrate seepage to groundwater, according to present knowledge. European and Flemish targets for emission reductions aim to move towards the long-term objective not exceeding these critical load values. For Flanders, critical load values for both nitrogen (N) and potentially acidifying pollutants were calculated by Langouche et al. (2002) and discussed in Chapter 1 (Table 1.1 and Table 1.3). Based on modelled deposition values (OPS model, Janssen & Mensink 2002), the exceedances of the critical load values calculated by Langouche et al. (2002) were reported in Overloop et al. (2006) and Van Avermaet et al. (2006). It was found that in 100%

of the forests in Flanders, biodiversity is threatened and soils are acidified as a consequence of elevated levels of N respectively potentially acidifying deposition. For protecting plant roots from damage and groundwater from nitrate pollution, the situation is less dramatic (Table 1.1, Table 1.3). Approximately 60% of all forest complexes in Flanders are smaller than 10 ha, indicating considerable fragmentation, and consequently a high forest edge/forest interior ratio. Several studies (including this thesis) report that forest edges act as 'hotspots' for N and potentially acidifying deposition, showing up to fourfold increases in the rate of atmospheric delivery compared with nearby areas without edges (Weathers et al. 2001).

If measures to control and reduce atmospheric N and potentially acidifying deposition are based on average deposition fluxes within forest stands, deposition reductions will not be enough to prevent adverse effects. We therefore calculated the relevance of considering the higher deposition load in forest edges for calculating the exceedance of critical load values for N and potentially acidifying deposition.

We used data of calculated critical load values for Flanders (Langouche et al. 2002), the forest mapping of Flanders, and averaged throughfall + stemflow deposition values in representative forest ecosystems in Flanders (ICP-forest monitoring plots, Genouw et al. 2003). The average exceedance of the critical load values for protecting forests from root damage was about 23% higher (0.79 $keq.ha^{-1}.yr^{-1}$) when elevated deposition fluxes in forest edges were considered (Fig. 4.2). Considering the critical load for protecting biodiversity from N deposition, it was calculated that the exceedance values were underestimated by 0.61 $keq.ha^{-1}.yr^{-1}$ (31%) (Fig. 4.2).

It was certainly not the aim to give an exact assessment of the error on the current calculations. We only intended to argue, by means of a simple calculation, that taking into account the spatial variability in depositions caused by, for example, forest edge effects, is relevant when critical load exceedances are calculated.

8.4 Do vegetation (and in particular forest) types differ in throughfall deposition of inorganic nitrogen and sulphur?

Vegetation types differ significantly in throughfall deposition quantities of inorganic nitrogen (N) and sulphur (S). Throughfall deposition is the net result of the interaction between atmosphere and forest canopy. As was explained in Chapter 1, the quantity of dry deposition

(gasses and particles that are directly deposited from the atmosphere onto vegetation), is strongly determined by the roughness of the deposition surface.

Chapter 5 of this study shows that heathland vegetations, which have a smaller height and surface roughness length compared to forests, receive significantly less nitrogen and sulphur throughfall deposition than forests. Furthermore, coniferous forests where found to receive higher N and S throughfall deposition compared to deciduous forests (Chapters 5, 6 and 7). This higher throughfall deposition of N and S in the coniferous plots is caused by higher dry deposition of gases and aerosols from the atmosphere. Since we did not study the possible causes in detail, we attribute this higher throughfall deposition fluxes in coniferous forest to a complex of several factors favouring dry deposition: the pine forests (1) are characterized by the highest trees and stand volume, (2) have evergreen foliage and (3) in case of the *Pinus nigra* plots a higher LAI compared to deciduous forests,. These characteristics have been described to be usually valid when deciduous and coniferous forest types are compared on similar soils (Augusto et al. 2002). In addition, the leaf shape affects the amount of ions deposited. Leaves with long narrow shape (needles) are according to Woodcock (1953 in Smith 1981) more efficient in salt accumulation than circular ones.

8.5 Is this forest type effect on nitrogen and sulphur throughfall deposition a general finding or coincidentally related to the studied forest types and emission regimes?

In the study region 'De Zwarte Beek', a part of Flanders where emission sources of NH_3 are hardly present within a radius of 10 km, the yearly nitrogen (N) throughfall depositions were on average 40% lower in deciduous forests (*Quercus robur* and *Betula pendula*) compared to coniferous forests (*Pinus sylvestris* and *Pinus nigra*) (see Chapter 5). Sulphur (S) throughfall deposition was on average 30% lower in deciduous forests compared to coniferous forests. In the study region 'Merksplas', where a high concentration of intensive livestock husbandry causes high NH_3 emissions, throughfall deposition of inorganic N in a *Betula pendula* forest was found to be 3.5 times lower than in a *Pinus nigra* plantation (Chapter 6). The observed higher throughfall depositions of inorganic N and S in coniferous forest stands were found in both the growing and the dormant season and account for regions with both relatively low and high deposition load.

Based on these two case-studies, it is very difficult to generalize our findings for other comparisons of coniferous and deciduous forest types, since the relative differences in throughfall deposition between the two forest types are highly dependent on the observed tree species, stand age, the performed forest management and the deposition load. In regions with medium to high deposition load, a comparison of e.g. *Betula pendula* and *Pinus nigra* will in theory generate higher differences than a comparison of e.g. *Betula pendula* and *Pinus sylvestris* or a comparison of *Picea abies* and *Fagus sylvatica,* because of the more profound differences in LAI, stand density and tree height between *Betula pendula* and *Pinus nigra*. Highly dense stands (a result of e.g. neglected thinning management) characterised by a high LAI will theoretically be subject to higher dry deposition quantities than forest stands with a small canopy density, although the latter can also experience high in-canopy wind speeds that stimulate dry deposition (Grace 1983).

Since the complexity of this matter, generalisation of our results was very difficult and somewhat speculative. Therefore we checked the scientific literature for other case studies on throughfall and seepage fluxes in comparable stands of coniferous and deciduous forest types (Chapter 7). The performed meta-analysis confirmed our general differentiation between coniferous and deciduous forest stands: coniferous forest types annually intercept more atmospheric N (on average about 2 times) and S (on average about 1.7 times) than deciduous forests at the same site and under the same climatic conditions and deposition load.

An interesting finding was furthermore that the higher the deposition load, the greater the difference in filtering capacity between coniferous and deciduous forest. Therefore, it can be expected that coniferous forests filter more efficiently in regions with high air concentrations of inorganic N and S than in regions with lower concentrations, and therefore receive higher quantities of inorganic N and S to the forest floor.

8.6 Do vegetation (and in particular forest) types differ in ion seepage quantities?

Linking vegetation types to ion seepage is less univocal than to throughfall deposition, since in ecosystems that are not yet nitrogen (N) saturated (for definitions of this concept see § 1.2.1), almost all anthropogenic N is used for plant and microbial growth as well as for accumulation in biomass and soil organic matter, especially in the forest floor. At some point, however, the input of N begins to exceed the abiotic and biotic demands for N within the

ecosystem. The system is predicted to lose its 'nitrogen retention capacity'. As this capacity is exceeded, excess N can be lost from the system via seepage water and gas fluxes.

All studied vegetation types were originally N limited, so without any significant seepage of nitrate. We assume that ecosystems are N saturated when nitrate seepage under the rooting zone occurs at a significantly higher rate than background levels (5 kg (357 mol) $N.ha^{-1}.yr^{-1}$), and during the entire year. It must, however, be clear that seepage quantities under the rooting zone of forests are not necessarily the quantities reaching the groundwater (see also §8.7)..

We did not study fluxes of N trace gas emissions, but the literature indicates that these losses are small compared to rates of N seepage (e.g. Butterbach-Bahl et al. 1998). However, it is interesting to report that in a recent study by Pilegaard et al. (2006) in 15 forest ecosystems across Europe, NO emissions were found to be significantly higher in coniferous compared to deciduous forests and significantly correlated to the nitrogen deposition load. In deciduous forests, NO emissions were found to be very low. On the contrary, N_2O emissions were slightly higher in deciduous compared to coniferous forests and no correlation with N deposition was found. The differences in nitrogen emissions between forest types were partly explained by differences in N deposition rates and partly by characteristics of the litter layer and soil. NO was mainly derived from nitrification, whereas N_2O from denitrification reactions. In general, soil moisture is lower at coniferous sites, and the litter layer of coniferous forests is thick and more aerated, favouring nitrification and thus release of NO. Conversely, the higher denitrification rates in deciduous than coniferous forests are due to a more compact and moist litter layer leading to N_2O production and NO consumption in the soil.

In the study region 'De Zwarte Beek', a region with relatively low emissions of NH_3, NO_x and SO_2, only appreciable aseasonal nitrate and sulphate seepages under the rooting zone were detected in the stands of *Pinus nigra*, while at the study site 'Merksplas', with a high concentration of NH_3 sources, nitrate seeped under both the *Betula pendula* and the *Pinus nigra* forest. Nitrate seepage under *Pinus nigra* was more than twice the seepage amount under *Betula pendula*. The *Pinus nigra* stand still retained 40% of the nitrogen flux to the forest floor, while in the *Betula pendula* stand almost the same nitrogen amount seeped as the incoming nitrogen throughfall flux. We hypothesized in chapter 6 (§6.5.6) that this difference is caused by the fact that the *Pinus nigra* plot is still accumulating litterfall and thus also nutrients and carbon in its forest floor. So on a yearly basis, not all nitrogen and other nutrients required for plant uptake are released from the litter layer (so no steady state) and

additional nitrogen has to be taken up originating from atmospheric deposition. It might be possible that the birch plot, because of the higher nutrient turnover in its litter layer, more bends to this steady state phase. The amount of nitrogen coming from the birch leaves might even be complemented by nutrient release from the former built-up forest floor under *Pinus sylvestris*.

As for throughfall deposition, we also searched for literature comparing ion seepage quantities in comparable coniferous and deciduous forest stands. In all stand comparisons included in the meta analysis, except one (Von Wilpert et al. (2000), ref. 37 in Table 7.1), NO_3^- seepage under coniferous forest was higher than under deciduous forest, although the differences between forest types were highly variable. The higher deposition load of SO_4^{2-} in coniferous forest stands was also reflected in a significantly higher seepage to the groundwater. The differences between coniferous and deciduous forests in SO_4^{2-} seepage fluxes were not that variable as for nitrate (see the smaller confidence interval for SO_4^{2-} in Table 7.3), and 3 out of the 11 studies reported higher SO_4^{2-} seepage under deciduous forests.

We found a close relationship between seepage of sulphate on the one hand, and K^+, Ca^{2+}, Mg^{2+} and Al(III) on the other hand. The higher seepage flux of anions under conifers clearly involves a higher soluble cation fraction, making total cation seepage a function of total anion seepage. The forest soil is strongly impoverished because nitrate seepage is linked to losses of so-called base cations, which reduces the buffering potential of the soil.

8.7 Risks and consequences of acidification and eutrophication for the observed forest ecosystems

The tree species in the studied forest ecosystems (*Pinus* spp., *Betula* spp. and *Quercus* spp.) are usually believed to be acid adapted (Zhong et al. 2003, Weber-Blaschke et al. 2002). Also the potential or natural plant species diversity is relatively low under pine plantations and oak/birch forests, although the latter have a somewhat higher number of bryophyte species (De Keersmaeker *et al* 2001, Dirkse & Martakis 1998). Furthermore, the soils in these areas have been acidified and have lost most of their buffering capacity throughout centuries of exhaustive agricultural use. Between 1250 and 1850 the prevailing farming system on sandy soils was more or less intensive sod cutting management (Bastiaens & Verbruggen 1996, Spek et al. 2004). The vegetation and organic topsoil were periodically removed from large areas of heathlands and degraded woodlands. Additionally large sheep herds grazed the

heathlands. This resulted in strong podzolisation and loss of soil fertility on the heathlands (Geudens et al. 2006). So taking into account these historically developed acid sandy soils and their naturally poor ecosystems in terms of biodiversity, one might question whether additionally nitrogen (N) and potentially acidifying deposition are still a big threat to the ecosystem.

For Dutch ecosystems on sandy soil, although typically poor in herbal biodiversity, changes in both herbal vegetation and mycoflora were observed due to the eutrophying effect of atmospheric nitrogen deposition (Van Tol et al., 1998). Except in the case of lichens (Aptroot *et al* 1998), eutrophication as a consequence of atmospheric N deposition has not yet resulted in a loss of species. On the contrary, total plant species abundance in Dutch forests has increased over the last decades (Samsen 1995 in Dirkse & Martakis 1998, van Dobben et al. 1994). However, significant effects were reported for the abundance of N-intolerant species (especially lichens of the genus *Cladonia*) that strongly declined, while N-demanding plant species abundance increased: *Deschampsia flexuosa, Molinia caerulea*, *Corydalis claviculata*, *Rubus sp.* and *Dryopteris dilatata*. In Flanders, 40% of our nitrophobe species are reported to be endangered (Overloop et al. 2004). Over the period 1960-1980, Arnolds (1988) showed a general decline of ectomycorrhizal fungi in the Netherlands, e.g. of yellow chanterelle (*Cantharellus cibarius)*. Meanwhile parasitic and saprophytic fungi tended to increase (Arnolds & Jansen 1992). Taylor et al. (2004) furthermore found that diversity of ectomycorrhizal fungi in spruce and beech stands decreased from North Europe to Central Europe, along with increasing atmospheric N deposition.

Besides the acid and nitrophilic conditions, it should be mentioned that in the studied forest types in this thesis, also forest age and past management affect the herbal biodiversity. Maturation of the forest floor and sustainable management practices (especially as rotations are lengthened, large clear cuts are avoided and deep soil preparation at the time of regeneration, litter raking and sod cutting stops and homogenous plantations will be changed in mixed species forests) will certainly improve the conditions for higher biodiversity, particularly for moss species (Van der Werf 1992; Geudens et al. 2006).

In all studied forests, significant seepage of so-called base cations and aluminium was reported causing a strong impoverishment and a reduction in buffering potential of the soil. Besides the reduced buffering potential, enhanced solubility of e.g. aluminium and also of manganese (not measured within the scope of this thesis) has a possible toxic influence on the vegetation. The resulting toxicity is element specific: the toxicity of manganese, which is an

element essential to plant growth, appears to be due to a nutrient imbalance brought about by abnormal accumulation in plant tissue (Sumner et al. 1991). By contrast, the effect of aluminium toxicity appears to be largely manifested as a pronounced malformation and malfunction of the root system of several tree species (Weber-Blaschke et al. 2002, Sumner et al. 1991) and vascular plants (e.g. Falkengren-Grerup and Tyler 1993). No research is however available specifically for the tree and vascular species in the observed forest types in Flanders under our specific conditions of high atmospheric depositions and naturally acid soils. Besides the direct toxic effect on roots, high aluminium concentrations in the soil solution can lead to worsening Ca^{2+} and/or Mg^{2+} supplies for the tree assimilatory tissues (Alewell et al. 2000, De Wit et al. 2001, Hruska et al. 2001, Weber-Blaschke et al. 2002) because of antagonistic effects between aluminium on the one hand and Ca^{2+} and Mg^{2+} on the other hand. Furthermore, Walters and Joergensen (1991), Raubuch and Beese (1993) and Zhong and Makeschin (2003) furthermore showed soil microbial biomass to be negatively correlated with soil exchangeable aluminium and positively with exchangeable calcium. From these studies it can be concluded that acid soil conditions are disadvantageous for soil microbial activities and soil biodiversity (Zhong and Makeschin 2003).

The observed seepage of nitrate, sulphate and aluminium to groundwater includes furthermore a risk for distribution of these ions via the waterflux to groundwater and surface water or to other groundwater influenced terrestrial and aquatic ecosystems. This risk for contamination highly depends on the local groundwater flow and the capacities for denitrification. There are three fates for nitrate in groundwater (Galloway et al. 2003): (1) accumulation, (2) conversion to gas or (3) distribution to other systems through hydrologic pathways. Accumulation of nitrate in groundwater seems not to be a major sink, since in regions of intense agricultural activity in Europe and the United States average nitrate accumulation rates in groundwater amount to at most a few percent of the N inputs (Howarth et al. 1996, in Galloway et al. 2003). N can be removed from groundwater both through denitrification to N_2, NO or N_2O and through losses of nitrate to surface waters. The relative contribution of both processes is however site dependent, as is the residence time in groundwater reservoirs (Nolan 1999, Groffman et al. 1998, both in Galloway et al. 2003).

8.8 Does a large-scale change in forest type negatively affect air quality and neighbouring ecosystems?

A frequently reoccurring concern regards the unknown impact of forest conversions on pollutant concentrations in the atmosphere and the eventual transport of pollutants to other ecosystems. Coniferous forest plots filter more pollutants from the atmosphere compared to deciduous forests. Does this filtering capacity contribute in terms of air purification considering (1) the high pollutant concentrations in the air and (2) the small surface area (1450 km²) of forests in Flanders? Calculations by Gielis and Wuyts (unpublished results) revealed that to purify the atmosphere from its current potentially acidifying emissions (10141 million $eq.yr^{-1}$), and assuming a value for throughfall deposition of e.g. 5000 $eq.ha^{-1}.yr^{-1}$ in coniferous forest, Flanders needs a forest surface area of 20280 km², or 150% of its total surface area (13522 km²).

So our coniferous forests do not contribute significantly to the achievement of clean air targets. What's more, pollutants are transferred from the atmosphere to the soil and groundwater, elevating the risk for the contamination of groundwater and other ecosystems, dependent on the local groundwater flow and the capacities for denitrification.

8.9 Overall conclusion with respect to forest ecosystem recovery

Depositions of nitrogen (N) and sulphur (S) did fall back during the past decade because of efforts made by both the industrial and the agricultural sector. The question rises to what extent Flemish and foreign policy makers can and will be prepared to enforce additional emission reductions. As can be seen from Chapter 1, we are far from achieving the long-term targets, and drastic emission reductions will be necessary to protect different vegetation types on the long term.

The previous and actual high rates of N and potentially acidifying depositions certainly raise questions about the time scale of soil and forest ecosystem recovery. As has been mentioned in Chapter 1, recent papers highlight that the deterioration of the soil environment with respect to acid stress and reduced so-called base cation availability has continued in spite of deposition reductions. Besides the too high depositon load of N and potentially acidifying pollutants, the release of SO_4^{2-} formerly stored in the soil is often an additional cause for this delay in the recovery of the soil solution. Clean-rain treatments (NITREX, EXMAN) in forest ecosystems highlighted a decrease in nitrate seepage within few years, while sulphate fluxes

remained high. The storage of both inorganic and organic N in the forest floor is, however, so high that each disturbance of the stand structure will lead to nitrate mobilization (Von Wilpert et al. 2000) generating a time-lag on the expected positive impact on nitrate concentrations in the soil solution.

This thesis confirms that converting coniferous *Pinus nigra* and *Pinus sylvestris* plantations into deciduous forests with *Betula pendula* and *Quercus robur* certainly diminishes the input of potentially acidifying and N deposition and consequently soil acidification and nitrate seepage to groundwater. Our results can furthermore be related to a recent analysis of Reich et al. (2005), who studied the effect of different tree species through litter quality on soil properties. Species-specific litter Ca^{2+} concentration appeared to be a key driver of decadal-scale change in soil properties, especially those related to acidity and cation biogeochemistry, but also those related to C and N contents and ratios in the forest floor and mineral soil horizons. Consequently, tree species rich in calcium were associated with increased native earthworm abundance and diversity, as well as increased soil pH, exchangeable calcium, percent base saturation and forest floor turnover rate (Reich et al. 2005). This was the case for both *Betula pendula* and *Quercus robur* compared to *Pinus nigra* and *Pinus sylvestris* in the study of Reich et al. (2005) and *Betula pendula* to *Pinus nigra* in our comparative study (Chapter 6). It should be noted, though, that the study of Reich et al. (2005) was conducted on a richer loamy sand soil than the sandy soil type considered in this thesis.

Forest stands dominated by conifers are furthermore often associated with a higher degree of cycling of aluminium than comparable hardwood stands (David and Driscoll 1984, Bergkvist and Folkesson 1995, Binkley and Valentine 1991). Brandberg and Simonsson (2003) state that a change in tree species composition from pure spruce stands to mixed birch/spruce stands can have a rather large short-term effect on the exchangeable and complex-bound aluminium and iron in organic matter of the forest floor. Spruce plots had a significantly higher concentration of exchangeable and organically bound aluminium in the O horizon and a lower concentration of each of the cations K^+, Ca^{2+} and Mg^{2+}.

In this respect, it is interesting to mention that a change in tree species composition not univocally changes the soil pH in spite of changes in base saturation. The higher base saturation in mixed spruce/birch stands studied by Brandberg and Simonsson (2003) compared to pure spruce stands did not result in a higher pH because the higher base saturation in mixed stands was compensated for by the lower aluminium saturation. The hydrogen ion saturation, which controls the pH, remained more or less unchanged. Also Van

Ranst et al. (2002) expressed their concern for the aluminization of the organic complexes in the forest floor in coniferous stands on sandy soil.

From the quite rapid (30-40 years) change in soil chemistry found by Reich et al. (2005) and Brandberg and Simonsson (2003), we might believe that a vegetation change can possibly contribute to reverse the high soil acidity and low soil fertility observed in these types of acid soils. The higher calcium concentrations in birch leaf litter will certainly positively influence soil chemistry, which can attract microbiota and other soil organisms that can contribute to nutrient turnover in soils. However it can hardly be expected that earthworms will colonize these soil types on the short-term. Muys (1993) detected in this respect only negligible quantities of epigeic earthworms under *Pinus nigra* and *Pinus sylvestris* plantations on sandy soil.

Vegetation, soil organisms and soil chemistry are known to be co-developing components of terrestrial ecosystems (Muys et al. 1992, Van Breemen 1993, Berendse 1998, Binkley and Giardina 1998 and Ponge et al. 1999) which highlights the need to consider the role of tree species as 'ecosystem engineer' for ecosystem recovery. Converting coniferous to deciduous forest types will certainly generate positive effects, but the conversion in itself already implies risks for nitrate seepage. Insight in the impact of different forest development phases and the chosen management strategy is lacking.

In order to improve acid soil conditions, some researchers have proposed a liming practice (e.g. discussed in Kreutzer 1995) since liming stimulates soil mineralization and nitrification through an increase in soil pH and Ca^{2+} and Mg^{2+} availability (Martikainen 1984). The long-term effect of liming on the stability of the ecosystem as a whole is still a matter of question. After liming, soil pH initially increases but this only seems to be a short-term effect (in case of Zhong and Makeschin only 4 years). Furthermore, Kreutzer (1995) found liming to have significant negative impacts on soil fertility because of unusually large changes in micro-organisms, collembolae and earthworms, inducing too fast decay of the humus layer, increased seepage of nitrate and mobilization of metals. Also Zhong and Makeschin (2003) propose that converting coniferous forests might be a more rational option for improving soil quality in a sustainable way.

A recent study of Boxman and Roelofs (2006) compared the effect of three management strategies (liming: addition of $CaCO_3$ and $MgCO_3$, sod cutting: removing the litter and humus layer and fertilization: addition of K and Mg in sulphate form) on the soil solution chemistry of a Scots pine stand at both ambient and decreased N deposition level. After three years, a

reduced N deposition level had a significant positive impact on soil solution concentrations. The authors furthermore propose an additional sod cutting as the most favourable management option to reduce nitrate seepage to groundwater, while liming and fertilization had less positive impacts. Sod cutting certainly removes a significant amount of nitrogen from the ecosystem (2400 kg N) but also of K^+ (40 kg), Ca^{2+} (200 kg), Mg^{2+} (30 kg) and P (50 kg). Comparing these values with nutrient cycling through litterfall (performed for a Scots pine forest of the ICP monitoring plot at Brasschaat), these amounts account for 10 to 25 times this flux, indicating that sod cutting significantly reduces the phosphorus and base cation stock of the forest. The nutrient pool available in the forest floor is however a necessarily basis for long-term recovery of these ecosystems, since the soils are very acid and low in base saturation. Long-term research and an analysis of the total nutrient budget are necessary to get insight in the global effect of this type of management strategies.

It should furthermore be emphasized that also in the observed deciduous forest type at 'Merksplas' clear symptoms of a nitrogen surplus and soil acidification processes were apparent. Therefore, conversion of coniferous to deciduous forests may certainly not be seen as *the* solution for this environmental problem. Converting coniferous to deciduous forest is, however, expected to cause improvements since at the status of nitrogen saturation, the ecosystem is very responsive to changes in N deposition (Gundersen et al. 1998b). Reductions caused by a change in tree species composition will therefore generally result in a significant decrease in nitrate seepage, with an undefined time delay because of accelerated mineralization and nitrification at the time of conversion. It must, however, be clear that conversion of coniferous forest to any other vegetation type will only delay the onset of nitrogen saturation, as the source of the problem (high nitrogen emissions) is still occurring in large parts of Europe. A reduction of the emission loads is certainly a necessary condition for the sustainable protection of forests and other ecosystems.

9 Summary

Forests on sandy soil did clearly acidify during the past decades, what was found to be related to high depositions of potentially acidifying compounds to the forest floor. Several factors are a stimulus for high atmospheric deposition load in these types of forest ecosystems. (1) Forests are often situated in the vicinity of intensive livestock husbandry, which is the cause of high NH_3 emissions to the atmosphere (one of the most important potentially acidifying pollutants at the moment). (2) Forests are often very small, so have a high surface area of forest edge (this is on average the outer 50 m zone of the forest). Forest edges act as 'hotspots' for potentially acidifying and nitrogen (N) deposition and can show up to four-fold increases in the rate of atmospheric delivery compared with nearby areas without edge. (3) The prevailing forest types are plantations of coniferous tree species (*Pinus sylvestris*, *Pinus nigra*) which have, in comparison to other vegetation types, a higher filtering capacity due to their higher leaf area index and evergreen character.

Besides the acidification of the soil, clear symptoms of N saturation were found in several forest ecosystems. All studied vegetation types were originally N limited, so without any significant seepage of nitrate. At the moment, nitrate seepage occurs at a significantly higher rate than background levels (5 kg (357 mol) $N.ha^{-1}.yr^{-1}$), and during the entire year.

When forest ecosystems are nitrogen saturated - and it might be expected that, under the current load of N emissions, this will hold true for most ecosystems on the long-term - a close relationship exists between the level of N deposition input and the level of N seepage. This work clearly demonstrates that different forest types, in this thesis simplified as coniferous versus deciduous forest types, have different impacts on ion throughfall and seepage fluxes. Coniferous forests receive significantly higher amounts of N and sulphur (S) to the forest floor than deciduous forests. Consequently, more nitrate and sulphate, and also more so-called base cations and aluminium are seeped to the groundwater. Besides a higher pollution of ground and surface water with nitrate and sulphate, the related higher seepage of so-called base cations and aluminium furthermore accounts for a higher reduction of the soil buffering capacity.

From this work it can be concluded that converting coniferous *Pinus nigra* and *Pinus sylvestris* plantations into mixed species forests with *Betula pendula* and *Quercus robur* certainly diminishes the input of potentially acidifying and N deposition and consequently soil acidification and nitrate seepage to groundwater. But it should be emphasized that also in some of the observed deciduous forest types clear symptoms of a nitrogen surplus and soil acidification processes were apparent. Therefore, conversion of coniferous to deciduous forests might certainly not be seen as *the* solution for this environmental problem. Reducing the emission loads is a necessary condition for the sustainable protection of forest and other ecosystems on the long-term.

10 Samenvatting

De verzurende en vermestende deposities in Vlaanderen behoren tot de hoogste van Europa. Ten gevolge hiervan zijn de voorbije tientallen jaren onze bossen op zandbodem sterk verzuurd en vermest. Verschillende factoren spelen hierbij een rol. (i) Bossen op zandbodem zijn dikwijls gelegen in de dichte nabijheid van intensieve veeteeltbedrijven, welke verantwoordelijk zijn voor hoge emissies van ammoniakgas (een van de belangrijkste potentieel verzurende polluenten in Vlaanderen op dit moment). (ii) Deze bossen zijn dikwijls zeer klein en bezitten een hoog aandeel bosrandzone. Bosranden treden op als 'hotspots' voor atmosferische verzurende en vermestende polluenten waardoor ze dikwijls een viervoud van de polluenten ontvangen in vergelijking met het boscentrum. (iii) De dominante bostypes zijn plantages van de naaldboomsoorten grove en Corsikaanse den (*Pinus sylvestris* & *Pinus nigra*) welke in vergelijking met andere vegetatietypes zeer efficiënt atmosferische polluenten uit de lucht filteren ten gevolge van hun hoge bladoppervlakte en hun immergroen karakter.

Naast de vastgestelde bodemverzuring vonden we duidelijke aanwijzigingen dat vele bostypes stikstofverzadigd zijn. De bestudeerde vegetatietypes zijn van nature echter stikstofgelimiteerd en vertonen dus in oorsprong geen of nauwelijks uitspoeling van stikstof naar het grondwater. In diverse bosecosystemen wordt momenteel uitspoeling vastgesteld van significante hoeveelheden nitraat (hoger dan de achtergrondwaarde van 5 kg N ha^{-1} j^{-1}) naar het grondwater en dit gedurende het ganse jaar.

Wanneer bossen stikstofverzadigd zijn - en het valt te verwachten dat op termijn onder het huidige emissieregime de meeste van onze bossen dat zullen zijn – is de nitraatuitspoeling sterk gerelateerd aan de atmosferische stikstofdepositie. Binnen dit onderzoek werd duidelijk vastgesteld dat verschillende bostypes, hier gesimplificeerd als naaldbos en loofbos, een verschillende impact hebben op de hoeveelheden atmosferische polluenten die in het bos terechtkomen en uitspoelen naar het grondwater. Bodems onder naaldbos ondervinden een significant hogere depositielast van stikstof en zwavel dan bosbodems onder loofbos. Ten gevolge hiervan spoelen hogere hoeveelheden nitraat en sulfaat uit, wat pollutie van het

grondwater veroorzaakt, maar ook van aluminium, kalium, calcium en magnesium, wat o.a. de buffercapaciteit van de bodem tegen verdere protoneninput vermindert.

We concluderen uit dit onderzoek dat een omvorming van naaldboomplantages naar gemengd loofbos duidelijk de depositielast van stikstof en verzurende partikels vermindert en daardoor een minder grote bedreiging vormt voor verzuring en vermesting van het ecosysteem en vervuiling van het grondwater. Echter, ook in de bestudeerde loofboombossen werden duidelijke symptomen van stikstofverzadiging en bodemverzuring vastgesteld. Een bosomvorming zoals gepresenteerd in dit werk kan dan ook op geen enkele manier gezien worden als de hoofdoplossing voor het bestudeerde milieuprobleem. Reductie van de emissies, zowel nationaal als internationaal, is de enige weg naar een duurzame bescherming van bossen en andere vegetatietypes op lange termijn.

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Curriculum vitae

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Education

1991-1995:	M.Sc. in Bioscience engineering, Faculty of Bioscience Engineering, Ghent University, Belgium

Professional experience

November 1995-februari 1996

Teacher mathematics, biology and geography at the Sint-Vincentius-Institute at Dendermonde.

March 1996-September 2001

Research associate at the Laboratory of Forestry, Ghent University

- Maart 1996-februari 1997: Schade aan dennenbossen in het Vlaams Gewest door Sphaeropsis sapinea : Fytopathologische aspecten en relatie met omgevingsfactoren, studie in opdracht van AMINAL, afdeling Bos & Groen.

- Maart 1997-September 2001: Intensieve monitoring van het bosecosysteem in het Vlaamse gewest, studie in opdracht van AMINAL, afdeling Bos & Groen.

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Teaching assistent at the department Forest and Water management

Scientific publications

a. Publications in international journals with peer review cited in the Science Citation Index (SCI impact-factor in 2005)

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1. De Schrijver A., Nachtergale L., Roskams P. De Keersmaeker L., Mussche S. & Lust N. (1998). Soil acidification along an ammonium deposition gradient in a Corsican Pine stand in northern Belgium. Environmental Pollution 102, S1, 427-431. SCI=2.451
2. De Schrijver A., Van Hoydonck G., Nachtergale L., De Keersmaeker L., Mussche S. & Lust N. (2000). Comparison of nitrate leaching under silver birch (*Betula pendula*) and Corsican pine (*Pinus nigra ssp. laricio*) in Flanders (Belgium). Water, Air and Soil Pollution, 122, 77-91. SCI=1.258
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4. Mussche S., Samson R., Nachtergale L., De Schrijver A., Lemeur R. & Lust N. (2001). A comparison of optical and direct methods for monitoring the seasonal dynamics of Leaf Area Index in deciduous forests, Silva Fennica, 35(4): 373-384. SCI =0.622
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8. Mertens J., Vervaeke P., De Schrijver A. & Luyssaert S. 2004. Metal uptake by young trees from dredged brackish sediment: limitations and possibilities for phytoextraction and phytostabilisation. Science of the Total Environment, 326, 209-215. SCI =2.224

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10. Devlaeminck R., De Schrijver A. & Hermy M. 2005. Variation in throughfall deposition across a deciduous beech (*Fagus sylvatica* L.) forest edge in Flanders. Science of the Total Environment 337, 241-252. SCI =2.224

11. Luyssaert S., Staelens J. & De Schrijver A. 2005. Does the commonly used estimator of nutrient resorption in tree foliage actually measure what it claims to? Oecologia 144, 177-186. SCI I= 3.032

12. Staelens J., De Schrijver A., Van Avermaet P., Genouw G. & Verhoest N. 2005. A comparison of bulk and wet-only deposition at two adjacent sites in Melle (Belgium). Atmospheric Environment 39, 7-15. SCI =2.724

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22. Staelens J., De Schrijver A., Verheyen K. & Verhoest N.E.C. Spatial variability and temporal stability of throughfall water under a dominant beech (*Fagus sylvatica* L.) tree in relationship to canopy cover. Journal of Hydrology, in press. SCI =1.745

23. Staelens J., De Schrijver A., Verheyen K. & Verhoest N.E.C. Rainfall partitioning into throughfall, stemflow, and interception within a single beech (*Fagus sylvatica* L.) canopy: influence of foliation, rain event characteristics, and meteorology. Hydrological Processes, in press. SCI =1.336

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2. De Schrijver A., Staelens J., Van Hoydonck G., Janssen N., Mertens J., Gielis L., Wuyts K.., Geudens G., Augusto L. & Verheyen K. Vegetation type determines element throughfall deposition and seepage quantities. Submitted to Environmental Pollution. SCI =2.451
3. Staelens J., De Schrijver A., Houle D., Neirynck J. & Verheyen K. Quantifying atmospheric deposition onto a deciduous foest canopy using varying forms of the canopy budget method and an inferential technique. Submitted to Water, Air and Soil Pollution. SCI =1.258
4. Vandenbruwane J., De Neve S., De Schrijver A., Geudens G., Verheyen K. & Hofman G.. Laboratory comparison of ceramic and PTFE/Quartz suction cups for sampling inorganic ions in the soil solution. Submitted to Communications in Soil Science and Plant analysis. SCI =0.442
5. Vande Walle I., De Schrijver A., Samson R., Verheyen K. & Lemeur R. Carbon sequestration in the Aelmoeseneie experimental forest (Gontrode, Belgium) from 1991 to 2005. Submitted to European Journal of Forest Science (new journal, no SCI).
6. Wuyts K., De Schrijver A., Staelens J., Gielis L. & Verheyen K. Spatial patterns of throughfall deposition along a transect in a forest edge of silver birch and Corsican pine. Submitted to Canadian Journal of Forest Research. SCI =1.531

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Muys B., Van Calster H., Aerts R., Valckx J., De Schrijver A., Verheyen K. & Scheffer M. Soil acidification and restoration in temperate forest ecosystems: alternative stable

states and the role of keystone species and ecosystem engineers. Trends in Ecology and Evolution. SCI=14.864

b. Publications in international journals without SCI-impact factor

7. Nachtergale L., De Schrijver A., Troch P. & Lust, N. 1996. Possibilities for forest development in a zone of controlled inundation in Kruibeke (Eastern Flanders) by means of a geographical information system. Silva Gandavensis 61, 46-55.
8. De Schrijver A., Nachtergale L., De Temmerman L., Frechilla, J.M.F., Mussche, S. & Lust, N. 1997. Evolution of soil acidity during the past 36 years (1960-1996) under coniferous forest in a region of high livestock production in Flanders. Silva Gandavensis 62, 45-50.
9. Nachtergale L., De Schrijver A. & Lust N. 1997. Windthrow, what comes after the storm? Silva Gandavensis 62, 80-89.
10. Staelens J., De Schrijver A., Oyarzún C. & Lust N. 2003. Comparison of dry deposition and canopy exchange of base cations in temperate hardwood forests in Flanders and Chile. Gayana Botanica 60, 9-16.

c. Chapters in books

1. Vanongeval L., Coppens G., De Keersmaeker L, De Schrijver A., Mussche S. & Lust N. (1998). Vermesting. In: Verbruggen A. (red.): Milieu- en Natuurrapport Vlaanderen: thema's. MIRA-T 1998, Vlaamse Milieu Maatschappij, 191-206.
2. Mensinck C., Viaene P., De Keersmaeker L., De Schrijver A., Mussche S. & Lust N. (1998). Verzuring. In: Verbruggen A. (red.): Milieu- en Natuurrapport Vlaanderen: thema's. MIRA-T 1998, Vlaamse Milieu Maatschappij, 207-220.
3. Mensinck C., Viaene P., De Keersmaeker L., De Schrijver A., Mussche S. & Lust N. (1998). Verzuring. In: Verbruggen, A. (Ed.): MIRA-T 1998. Wetenschappelijk rapport, 13, 1-39.
4. Mensink C., Colles A., De Schrijver A., Hendriks J. & Meykens J. 2000. Verzuring. In : Van Steertegem M. (Ed.). MIRA-S 2000. Milieu- en Natuurrapport Vlaanderen: Scenario's. Garant, Leuven, Apeldoorn, p. 383-394.

5. Van Gijseghem D., De Schrijver A., Van Hoydonck G. & Lust N., Mensink C. 2000. Vermesting. In: Van Steertegem M. (Ed.). MIRA-S 2000. Milieu- en Natuurrapport Vlaanderen: Scenario's. Garant, Leuven, Apeldoorn, p. 367-382.

6. Dumortier M., Schneiders A., Boeye D., De Schrijver A., De Keersmaeker L., Martens K, Vervaet H., Van der Welle J. & Van Damme S. 2001. #5.3 Vermesting. In: Kuijken, E., Boeye, D., De Bruyn, L., De Roo, K., Dumortier, M., Peymen, J., Schneiders, A., van Straaten, D. & Weyembergt, G. (Eds.). Natuurrapport 2001. Toestand van de natuur in Vlaanderen: cijfers voor het beleid. Mededeling van het Instituut voor Natuurbehoud. pp. 125-138.

7. Dumortier M., Schneiders A., Boeye D., De Schrijver A., De Keersmaeker L., Martens K, Vervaet H., Van der Welle J. & Van Damme S. 2001. #5.3 Vermesting. In: Kuijken E., Boeye D., De Bruyn L., De Roo K., Dumortier M., Peymen J., Schneiders A., van Straaten D. & Weyembergt G. (Eds.). Natuurrapport 2001. Toestand van de natuur in Vlaanderen: cijfers voor het beleid. Mededeling van het Instituut voor Natuurbehoud. pp. 125-138.

8. Van Gijseghem D., Dumortier M., Van Landuyt W., Goethals V., De Schrijver A., Mensink C. & Overloop S. 2001. Vermesting. In: Van Steertegem M. (Ed.). Mira-T 2001, p. 291-303.

9. Dumortier M., De Schrijver A., Denys L. & Van Reusel W. #20. Verzuring. In: Dumortier M., De Bruyn L., Peymen J., Schneiders A., Van Daele T., Weyembergh G., van Straaten D. & Kuijken E. (Eds.). Natuurrapport 2003. Toestand van de natuur in Vlaanderen: cijfers voor het beleid. Mededeling van het Instituut voor Natuurbehoud, Brussel. pp. 148-153

10. Dumortier M., Schneiders A., De Schrijver A., Kyramarios M., Van Daele T., Van Damme S. & Denys L. #19. Vermesting. In: Dumortier M., De Bruyn L., Peymen J., Schneiders A., Van Daele T., Weyembergh G., van Straaten D. & Kuijken E. (Eds.). Natuurrapport 2003. Toestand van de natuur in Vlaanderen: cijfers voor het beleid. Mededeling van het Instituut voor Natuurbehoud, Brussel. pp. 138-147.

11. Dumortier M., Genouw G., Neirynck J., Overloop S., Van Avermaet P., De Schrijver A., Devlaeminck R. # 19. Verzuring. In: Kuyken E., Boeye D., De Bruyn L., De Roo K., Dumortier M., Peymen J., Schneiders A., van Straaten D. & Weyembergt G.

(Eds.). Natuurrapport 2005. Toestand van de natuur in Vlaanderen: cijfers voor het beleid. Mededeling van het Instituut voor Natuurbehoud, Brussel., pp.204-210.

e. Editor

1. De Schrijver A., Kint V. & Lust N. 2002. Comparison of ecosystem functioning and biogeochemical cycles in temperate forests in southern Chile and Flanders. Gent, Uitgeverij Academia Press, 2002, 129 pp. ISBN 90 382 0393 4.
2. De Schrijver A., Kint V., Geudens G. & Lust N. 2002. Bosomvorming. Noodzaak en praktijk van een effectgerichte maatregel tegen verzuring en vermesting van bossen op zandgrond. Gent, Uitgeverij Academia Press., 2002, 93pp. ISBN 90 382 0393 4.

f. National publications without peer-review

1. De Schrijver A., Nachtergale L. & Lust N. 1998. Windval: wat gebeurt er na de storm? Groene Band104, 1-30.
2. De Keersmaeker L., De Schrijver A., Nachtergale L., Mussche S. & Lust N. 1998. Evaluatie van bosomvorming als effectgerichte maatregel tegen verzuring en vermesting van bossen. Groene Band 105, 31 p.
3. Campforts E., De Schrijver A. & Lust N. 2001. Verzuring van zandige bosbodems in Vlaanderen tijdens de periode 1950 – 2000. Groene Band 114, 22-37.
4. Geudens G., De Schrijver A. & Nachtergale L. 2004. Adelaarsvaren: ecologie. Bosrevue 10, 5-7.
5. De Schrijver A., Geudens G. & Nachtergale L. 2005. Adelaarsvaren: perceptie door Vlaamse beheerders. Bosrevue 12, 12-14.
6. Geudens G., Verheyen K., De Schrijver A. & Nachtergale L. 2005. Adelaarsvaren: het beheer. Bosrevue 14, 6-9.
7. Van Herzele A., Geudens G., De Schrijver A. & Verheyen K. 2006. Hoe eigenaars warm maken voor duurzaam bosbeheer. Science Connection 12, 28-29.
8. Verheyen K., De Schrijver A., Wuyts K., Gielis M., Van Gossum P. Geudens G., Van Herzele A., De Boever L., Vanhellemont M. (2007). Van dennenplantages naar een

beloofd land?! Theoretische en praktische aspecten van bosomvorming. Silva Belgica, 114: 20-26.

g. Abstracts of presentations at scientific congresses

1. De Schrijver A., Nachtergale L., Roskams P., De Keersmaeker L., Mussche S. & Lust N. 1998. Soil acidification along an ammonium deposition gradient in a Corsican Pine stand in northern Belgium. In. Van der Hoek K.W., Erisman J.W., Smeulders S., Wisniewski JR. & Wisniewski J. (Eds.). Proceedings of the First International Nitrogen Conference, Noordwijkerhout (The Netherlands), pp. 427-433
2. De Schrijver A., Van Hoydonck G., Nachtergale L., Mussche S., De Keersmaeker L., Lust N. 2000. Impact of forest conversion on nitrogen saturation in Flanders. In : Hasenauer H. (Ed.). Proceedings of the International Conference on Forest Ecosystem Restoration, Vienna (Austria), 10-12 April 2000, p. 77-84.
3. Van Hoydonck G., Cabus P., De Schrijver A., Verhoest N., Lust N., De Troch F. 2000. Modeling the impact of vegetation (heather, deciduous forest, coniferous forest) of infiltration areas on water quality in catchments. In : Hasenauer H. (Ed.). Proceedings of the International Conference on Forest Ecosystem Restoration, Vienna (Austria), 10-12 April 2000, p. 277-280.
4. De Schrijver A., Nachtergale L., Campforts E., Van Hoydonck G., De Keersmaeker L. & Lust N. 2002. Bosomvorming als effectgerichte maatregel tegen verzuring en vermesting van bossen op zandgrond. Uitgeverij Academia Press, Gent, p. 27-38.
5. De Schrijver A., Nachtergale L., Van Hoydonck G. & Lust N. 2002. Forest conversion as an effective measure to counteract soil acidification and nitrate leaching in forests on sandy soils. Proceedings of the Workshop held at Ghent University, Belgium, 17-19 September 2001. Uitgeverij Academia Press, Gent, p. 35-48.
6. Staelens J., De Schrijver A.., Oyarzún C. & Lust N. 2002. Comparison of dry deposition and canopy exchange of base cations in temperate hardwood forests in Flanders and Chile. International workshop on biogeochemical cycles in temperate forest ecosystems, 11-13 November 2002, Valdivia, Chile.
7. Van Hoydonck G., Delannoy G., De Schrijver A., Verhoest N., Lust N. & De Troch, F. 2002. Modellering van de impact van het vegetatietype (loofbos, naaldbos en heide)

in infiltratiegebieden op de waterkwaliteit in het stroombekken – Deposities en uitspoeling van nutriënten onder bos en heide in het stroombekken van de Zwarte beek. Uitgeverij Academia Press, Gent, p. 39-48.

8. Van Hoydonck G., De Lannoy G., De Schrijver A., Verhoest N., De Troch F. & Lust N. 2002. Deposition and leaching of nutrients under forest and heather in the catchment of the Zwarte beek. Proceedings of the Workshop held at Ghent University, Belgium, 17-19 September 2001. Uitgeverij Academia Press, Gent, p. 49-58.

9. De Schrijver A., Nachtergale L., Staelens J., Luyssaert S., De Keersmaeker L. & Lust N. 2003. Nitrogen saturated forest ecosystems: Forest conversion as an effective measure to counteract soil acidification and nitrate leaching. Bericht über den Workshop Integrierende Auswertung der Daten des Forstlichen Umweltmonitoring (Level I/II) vom 24.-26. Februar 2003 in Bonn-Röttgen. BMVEL in Zussamenarbeit mit der Bund-Länder-Arbeitsgruppe 'Forstliches Umweltmonitroging' und dem IFOM Project des BMBF, pp. 33-37. Invited speaker.

10. De Schrijver A., Nachtergale L., Staelens J., Luyssaert S., De Keersmaeker L. & Lust N. 2003. Nitrogen saturated forest ecosystems: Forest conversion as an effective measure to counteract soil acidification and nitrate leaching. Poster abstract in: International symposium on Forest ecosystem and landscape research: scientific challenges and opportunities, 25-27 June 2003, Tours, France.

11. Staelens J. & De Schrijver A. 2003. Spatial variability of throughfall water under beech (*Fagus sylvatica* L.). Poster abstract in: International symposium on Forest ecosystem and landscape research: scientific challenges and opportunities, 25-27 June 2003, Tours, France.

12. Staelens J., De Schrijver A., Oyarzún C. & Lust N. 2003. Atmospheric nitrogen input in temperate hardwood forests in Chile and Flanders. Abstract of oral presentation in: International workshop on risk assessment of agricultural intensification on N deposition on pristine forests and plantations in southern Chile, 22-26 April

13. Staelens J. & De Schrijver A. 2004. Spatial variability of throughfall water under beech (*Fagus sylvatica* L.). Poster abstract in: Proceedings of the 10th PhD Symposium on Applied Biological Sciences. Communications in Agricultural and Applied Biological Sciences 69, 273-274.

14. Staelens J., De Schrijver A. & Van Avermaet P. 2005. A comparison of bulk and wet-only deposition in Flanders (Belgium). Poster abstract in: Hůnová, I. (ed.). Conference abstracts of Acid Rain 2005, 7th International conference on acid deposition, 12-17 June 2005, Prague, Czech Republic, p. 130. 2003, Ghent, Belgium

h. Vulgarizing publications

1. De Schrijver A., Nachtergale L. 1996. Windval, een bos behoeft geen stervensbegeleiding. De Boskrant, 3, 80-84.
2. De Schrijver A. 2001. Verzuring in Vlaanderen: geen vuiltje aan de lucht? De Boskrant, 31 (5) p.26.
3. Meiresonne L., De Schrijver A. 2003. Populieren en stikstof. Bosrevue 2(5), p.10.
4. De Schrijver A. 2000. Het bos en MAP2. De verantwoordelijkheid van de bossector in de vermestingsprolematiek. De Boskrant, 2-5.
5. De Schrijver A. 2003. Zure regen hoeft niet nat te zijn. Alarm voor bodems en bossen. Onze bossen worden armer. De Verrekijker. Verzuring in Vlaanderen, 7-16.
6. Wetenschapsbijlage De Standaard 16 mei 2003: 'Zure regen zorgt voor oprispingen' door Kim De Rijck

   Opiniebijdrage De Standaard 16 mei 2003: Reactie van UGent en Instituut voor Bosbouw & Wildbeheer: 'Zure regen bestaat wel'.

   Dit betrof reacties op artikels in de Standaard, De Gentenaar & Het Nieuwsblad (13& 14 mei 2003) met als titels: 'Zure regen bestaat niet', 'Bossen zijn belangrijke bronnen van vervuiling' en 'Debat over zure regen barst los'.

i. Scientific reports

1. Brauwers C., De Schrijver A., Poppe J. & Lust N. 1997. Schade aan dennenbossen in het Vlaams Gewest door Sphaeropsis sapinea : Fytopathologische aspecten en relatie met omgevingsfactoren. AMINAL, Laboratorium voor Fytopathologie & Laboratorium voor Bosbouw, RUG, 119 p.
2. De Schrijver A. & Lust N. 1998. Deelaspecten van de intensieve monitoring van het bosecosysteem in het Vlaamse Gewest - meetjaar 1997. Universiteit Gent, Laboratorium voor Bosbouw. Eindverslag 1998, contract nr. IBW/1/1998, 120p.

3. De Schrijver A. & Lust N. 1999. Deelaspecten van de intensieve monitoring van het bosecosysteem in het Vlaamse Gewest, Eindverslag 1999. Contract nr. IBW/1/1999, Laboratorium voor Bosbouw, Universiteit Gent, 65p.
4. De Schrijver A. & Lust N. 2000. Deelaspecten van de intensieve monitoring van het bosecosysteem in het Vlaamse Gewest - meetjaar 1999. Universiteit Gent, Laboratorium voor Bosbouw. Eindverslag 2000, contractnr. IBW/1/2000. 147 p.
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6. Nachtergale L., De Schrijver A. & Lust, N. 2002. Deelaspecten van de intensieve monitoring van het bosecosysteem in het Vlaamse Gewest, meetjaar 2001. Project in opdracht van de Vlaamse Gemeenschap, IBW/1/2002, 2002.60.B.L., 60 p.
7. Verstraeten W.W., Minaert M., Pieters M., Goessens S., Hubrechts L., De Schrijver A., Samson R., Feyen J., Deckers J. Muys B., Lust N., Lemeur R., Van Slycken J. & Devos B. (2001). Kwantitatieve analyse van de verdamping van bossen in vergelijking met weide en akkerland. Eindverslag VLINZ 99/06, studie uitgevoerd voor rekening van de Vlaamse Gemeenschap binnen het kader van het Vlaams Impulsprogramma Natuurontwikkeling in opdracht van de Vlaamse minister bevoegd voor natuurbehoud, 196p.
8. Verstraeten A., De Bruyn L., De Keersmaeker L., Vandekerkhove K., Smets K., D'Havé H., Lust N., De Schrijver A. & Willems L. 2004. Evaluatie van beheermaatregelen om de ecologische waarde van populierenbossen te optimaliseren. PBO-project 99A/35/69. 275 pp.

Active participation in congresses, symposia and workshops

1. The N-conference, 23-27/3/1998 te Amsterdam *Poster and short oral presentation*: Soil acidification along an ammonium deposition gradient in a Corsican pine stand in Northern Belgium
2. Watershed Management Conference, The Science of Managing Forests to Sustain Water Resources, 8-11/11/1998 te Sturbridge, Massachussets (US). *Oral and poster*

presentation: Forest conversion as an effective strategy for protecting drinking water pools against nitrate pollution

3. Conference on Forest Ecosystem Restoration, 10-12/4/2000 te Wenen. *Oral presentation*: Impact of forest conversion on nitrogen saturation in Flanders.
4. Studiedag: "Bosomvorming: noodzaak en praktijk van een effectgerichte maatregel tegen verzuring en vermesting". 29/11/2001 te Gent. *Voordracht*: Bosomvorming als effectgerichte maatregel tegen verzuring en vermesting van bossen op zandgrond.
5. Workshop: 'Comparison of ecosystem functioning and biogeochemical cycles in temperate forests in Southern Chile and Flanders'. 17-19 September 2001 at Ghent. *Oral presentation*: Forest conversion as an effective measure to counteract soil acidification and nitrate seepage to forests on sandy soils
6. Workshop: "Integrierende Auswertung der Daten des forstlicher Umweltmonitoring". 24-26 Februari 2003 te Bonn. *Oral presentation, invited speaker:* Nitrogen saturated forest ecosystems: Forest conversion as an effective measure to counteract soil acidification and nitrate leaching.
7. Conference: Towards the Sustainable Use of Europe's Forests. Forest Ecosystem and Landscape Research: Scientific Challenges and Opportunities. 25-27 June 2003 te Tours. *Poster presentation*: Forest conversion as an effective measure to counteract soil acidification and nitrate leaching
8. Studiedag: "Van dennenplantages naar een beloofd land: theoretische en praktische aspecten van bosomomvorming. 23/11/2006 te Hasselt. *Voordracht*: Scenario's voor bosomvorming op bestands- en boscomplexniveau.

Author/co-author of approved projects

1. Project ingediend bij Ministerie van de Vlaamse Gemeenschap - Departement Wetenschap, Innovatie en Media: Administratie Wetenschap en Innovatie: Programma Beleidsgericht onderzoek. Evaluatie van beheersmaatregelen om de ecologische waarde van populierenaanplantingen te optimaliseren
2. Project ingediend bij het Vlaams Impulsprogramma voor Natuurontwikkeling . Modellering van de impact van vegetatie (heide, loofbos, naaldbos) van infiltratiegebieden op de waterkwaliteit in stroombekkens

3. Project ingediend bij het Vlaams Impulsprogramma voor Natuurontwikkeling. Titel: Kwantitatieve analyse van de verdamping van bossen in vergelijking met weide en akkerland
4. Project ingediend bij Universiteit Gent, Bijzonder onderzoeksfonds: Bilaterale wetenschappelijke samenwerking. Comparison of ecosystem functioning and biogeochemical cycles in temperate forests in southern Chile and Flanders
5. Project ingediend bij Universiteit Gent, Bijzonder onderzoeksfonds: Bilaterale wetenschappelijke samenwerking. Risk assessment of agricultural intensification on nitrogen deposition on pristine forests and plantations in Southern Chile
6. Project ingediend bij Afdeling Land en Tuinbouw. Evaluatie van het programma voor plattelandsontwikkeling in Vlaanderen – maatregelen inzake bosbouw
7. Project ingediend bij DWTC: Second multi-annual scientific support plan for a sustainable development policy – SPSD II. Environmental, social and economic feasibility of forest conversion – FEFOCON.
8. Project ingediend bij AMINAL – Bos & Groen. Natuurvriendelijker exploitatiewijzen voor bossen op kwetsbare gronden
9. Project ingediend bij Universiteit Gent, Bijzonder onderzoeksfonds: Bilaterale wetenschappelijke samenwerking. Effect of fire damage on N and P losses from in *Araucaria araucana* ecosystems in Chile
10. Project ingediend bij Ministerie van de Vlaamse Gemeenschap – Afdeling Bos & Groen. Potentie van bosomvorming als effectgericht georiënteerde maatregel tegen bodemverzuring en eutrofiëring van bossen op zandgrond
11. Behaalde doctoraatsbeurs bij IWT (Karen Wuyts). De mitigerende werking van een aangepast bosrandbeheer op depositie, nitraatuitspoeling en bodemverzuring in bosecosystemen op arme zandgrond
12. Project ingediend bij de Vlaamse MilieuMaatschappij. Statistische verwerking van meetdata verzuring

Supervision of M.Sc. thesis students

1. Dossche Tomas, 1998. Ecologische effecten van bladstrooisel van loofboomsoorten op de ontwikkeling van recent beboste landbouwgronden (Mortagnebos – Zwevegem)
2. Van Hoydonck Gert, 1999. Evaluatie van bosomvorming als effectgerichte maatregel om verzuring en vermesting van bossen op zandgronden tegen te gaan.
3. Campforts Elke, 2001. Verzuring van zandige bosbodems in Vlaanderen in de periode 1950-2000.
4. Bauwens Bram, 2001. Stuurvariabelen voor vegetatiedynamiek in het meerdaalwoud (Vlaams Brabant) over de periode 1954-2000.
5. Janssen Noah, 2001. Studie van de nutriëntenflux in vochtige heide-ecosystemen in de Vallei van de Zwarte beek.
6. Leen Gielis, 2004. Studie van deposities en uitspoeling van polluenten in een gemengde verjongingsgroep onder scherm van grove den.
7. Ive Van Bouwel, 2004. Studie van deposities en uitspoeling van polluenten in de bosrand van een homogeen naaldbos.
8. Karen Wuyts, 2004. Studie van deposities en uitspoeling van polluenten in de bosrand van een gemengd loofbos.
9. Bart De Vos, 2006. Wortelbiomassa van *Salix viminalis* L. onder korte rotatie in relatie met bodemorganische koolstof (BOC)
10. Helena Bonne, 2007. Vergelijkende studie van kroonuitwisseling bij beuk (*Fagus sylvatica*), gewone es (*Fraxinus excelsior*) en zomereik (*Quercus robur*)